RADIATION TECHNOLOGY

IN FOOD, AGRICULTURE, AND BIOLOGY

CFTRI-MYSORE
5840
Radiation techno

13.50
20/12

Acc-No. 5840

1. radioactivity
2. radiation sources
3. living organisms
4- radiation processing

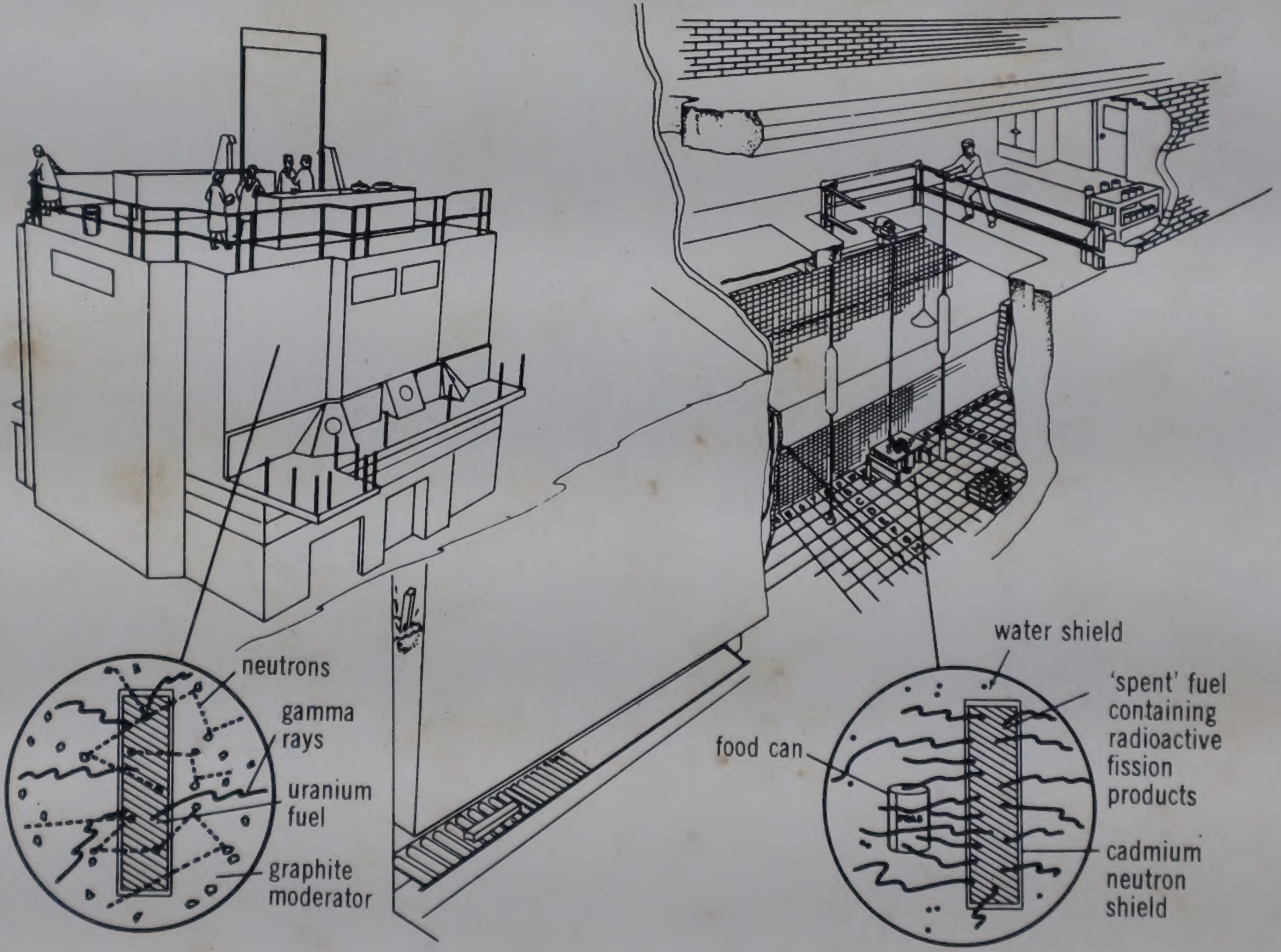
TZ48

Radiation Technology

in Food, Agriculture and Biology

other AVI books on food processing

Amerine and Cruess	The Technology of Wine Making
Desrosier	Technology of Food Preservation
Matz	Chemistry and Technology of Cereals as Food and Feed
Matz	Bakery Technology and Engineering
Merory	Food Flavorings Composition, Manufacture, and Use
Talburt and Smith	Potato Processing
Tressler and Joslyn	Chemistry and Technology of Fruit and Vegetable Juice Production
Tressler and Evers	Freezing Preservation of Foods Volume I Freezing of fresh foods Volume II Freezing of precooked and prepared foods


neutrons
gamma rays
uranium fuel
graphite moderator
water shield
food can
'spent' fuel containing radioactive fission products
cadmium neutron shield

Courtesy of QM. Food and Container Institute

A NUCLEAR REACTOR YIELDS POWER AND USEFUL BY-PRODUCTS OF RADIATION AND RADIOISOTOPES

Radiation Technology

in Food, Agriculture and Biology

---

by NORMAN W. DESROSIER, Ph.D.

Professor of Food Technology, Purdue University, Lafayette, Indiana. Formerly, Director, Food Radiation Preservation Division, Quartermaster Food and Container Institute for the Armed Forces, Chicago, Illinois, and member of "Atoms for Peace" Mission to Europe

and HENRY M. ROSENSTOCK, Ph.D.

Senior Radiation Scientist, William H. Johnston Laboratories, Inc., Baltimore, Maryland. Formerly, Research Physicist, Operations Research, Inc., Silver Spring, Maryland. Formerly Physical Chemist, Oak Ridge National Laboratory, Oak Ridge, Tennessee

WESTPORT, CONNECTICUT

THE AVI PUBLISHING COMPANY, INC.

1960

F8,3:6;R N60
~~K0~~

© *Copyright 1960 by*

THE AVI PUBLISHING COMPANY, INC.
Westport, Connecticut

REGISTERED AT STATIONERS' HALL
LONDON, ENGLAND, 1960

All rights reserved

1st Copy

5840

dt 17/4/64

CENTRAL FOOD TECHNOLOGICAL RESEARCH INSTITUTE * MYSORE *
LIBRARY

Printed in the United States of America
BY MACK PRINTING COMPANY, EASTON, PENNSYLVANIA

CFTRI-MYSORE
5840
Radiation techno..

Preface

The discovery of the chain reaction opened a new horizon of scientific endeavor. Nuclear fission became practically applied in weaponry; it is now yielding its power to more beneficial applications as a controlled source of energy for man's growing needs.

The purpose of this book is to bring together pertinent information from the physical and biological sciences and focus on some of the positive uses of atomic energy in the fields of food, agriculture and biology. Like fire, radiation can be either beneficial or harmful, and controlled applications may lead to most valuable results. For example, while it is relatively well known that high level irradiation can destroy a nutrient such as vitamin C, it may not be as widely recognized that vitamin C can be synthesized with radiation. Furthermore, the world-wide control of those parasites and food-poisoning organisms transmitted in our food and water supplies appears possible within presently existing radiation technology.

The health and welfare of mankind are intimately interwoven in the fabric called the economics of food distribution. The peaceful applications of atomic energy in the disinfection and stabilization of man's food directly involve the economics of food distribution. Radiation technology is beginning to yield direct contributions to the health and welfare of all men. The solution to widespread applications of atomic energy in the fields of food, agriculture and biology requires the best talents man can muster. The contributions to be made are most diverse in scope and nature. For this reason, a multitude of professional talents are required. There is need for biologists, food technologists, biochemists and agriculturists to yield the disinfected, stabilized foodstuffs. There is need for physiologists and nutritionists to establish that radiostabilized food is good for man to eat and is rewarding in yielding the required nutrients in proper combinations for buoyant health. There is need for chemists and container engineers to develop new protective packages for treated substrates to prevent reinfections. There is need for physicists and nuclear engineers to design processes for irradiation which are functional, do not induce worrisome radioactivity in treated materials or containers, yet are economical in application.

There is need for legislatures to yield laws to control this new process and its application. There is need for economists to consider the changing supply and demand and distribution channels for stabilized foods. There is need for food industrialists, and industrialists of allied fields to apply successfully the findings of this activity for the betterment of mankind.

This book was written with all these people in mind. Some of the information will be familiar to certain readers; others will desire that more detail should have been presented. It is a major purpose of this undertaking to present a background of information upon which one may reach decisions to launch programs and explore new applications. Man's creative ability has yet to be unleashed on this subject.

We perhaps can no more envision the uses of radiation in the next century than early man could conceive of present man carrying a package of matches in his pocket.

The next few years should find exploitation of the hundreds of millions of curies of radioactive materials which now silently decay away in caves and reservoirs, at great economic loss and expense to our society. Even if this presentation can but stimulate interest in this subject, it will have been sufficient reward for the undertaking.

We are indebted to Purdue University and the W. H. Johnston Laboratories, Inc., respectively for the opportunity to develop this manuscript.

We would like to acknowledge the assistance and guidance given to us by Dr. Donald K. Tressler and Dr. H. Elliott Foote of the Avi Publishing Company. Had their patience not exceeded our ineptness, this book might never have been completed.

Dr. Martin S. Peterson, Chief, Technical Services Office, QM. Food and Container Institute for the Armed Forces, Chicago, Illinois, has been most generous in aiding us to secure the many photographs from the Institute and its more than 100 contractors in radiation research. We also wish to acknowledge the information and photographs made available by Colonel Tyron Huber and Lt. Colonel L. Hirsch, Research and Development, Office of The Surgeon General, U. S. Army, and Office of Isotopes Development, Dr. Paul Aebersold, Director, U. S. Atomic Energy Commission and its contractors.

We would like to acknowledge the assistance given by the European Nuclear Energy Agency, and the European Productivity Agency of the Organization for European Economic Cooperation, Paris, France, and the atomic energy, agricultural and biological science groups in Austria, Belgium, Denmark, France, Greece, Iceland, Ireland, Italy, Norway, Portugal, Spain, Sweden, Switzerland, The Netherlands, Turkey, United Kingdom, West Germany and Yugoslavia for permitting us to repro-

duce portions of the information gained during a tour of duty with them on the Atoms for Peace Mission in 1958.

We would like to recognize the technical assistance given by former associates in the Radiation Preservation of Food Project of the U. S. Department of Defense: Mr. K. R. Clark, Lt. Col. George E. Danald, Mr. Maurice Drake, Dr. H. E. Goresline, Dr. A. W. Harvey, Dr. F. Heiligman, Dr. John Hilchey, Maj. R. Pomerantz, Mr. David Rest, Lt. Col. Robert Ryer III, Mr. Morris Simon, Dr. Ralph Siu, and Mr. George Tripp.

We are most appreciative of the following who reviewed the manuscript and suggested helpful improvements:

Dr. M. L. Fields, Professor of Food Technology, Purdue University; Dr. Jules Janick, Professor of Genetics, Purdue University; Dr. Ruble Langston, Professor of Plant Physiology, Purdue University; Dr. Charles Muller, Professor of Physical Chemistry, Purdue University; Dr. M. P. Plumlee, Professor of Animal Science, Purdue University; Dr. Thomas Plummer, Plant Physiologist, Purdue University; Dr. S. P. Sen, Plant Biochemist, Bose Research Institute, Calcutta, India; Dr. M. B. Wallenstein, consultant to the Associate Director for Chemistry, National Bureau of Standards, Washington, D. C.; Dr. Meinhard Zenk, Physiologist, University of Munich, Munich, Germany.

We are indebted to all researchers and teachers in many fields of knowledge touched on in this presentation. It is a portion of the cumulative knowledge of generations of contributors who developed our present understanding. We have attempted to bring this information together with as much visual material as practical and have drawn freely from many sources, which are cited in reference. While the horizon of information in radiation technology is to be found in the current literature, we hope that this book will be useful to the reader as background for future developments.

Recognizing that we, of necessity, had to cover many areas in the physical and biological sciences, and recognizing too, our personal limitations, the coverage of the literature was intended to be representative rather than comprehensive. Therefore, we sincerely hope that our sins of omission will be forgiven.

It is a pleasure to acknowledge the assistance given us by Ann Desrosier and Shirley Rosenstock during this undertaking. Without their patient encouragement we could not have reached this point.

NORMAN W. DESROSIER
HENRY M. ROSENSTOCK
West Lafayette, Indiana

January 4, 1960

Contents

CHAPTER PAGE

Part I. Introduction to Radiation and Its Effects

1. INTRODUCTION 3
2. NUCLEAR STRUCTURE AND RADIOACTIVITY 7
3. ATOMS AND MOLECULES 20
4. CHEMICAL REACTIONS INDUCED BY RADIATION 37
5. PHYSICAL EFFECTS OF RADIATION 46

Part II. Radiation Source Technology

6. RADIATION SOURCES 61
7. MEASUREMENT OF RADIATION DOSE 89

Part III. Interaction of Radiation with Living Organisms

8. RADIOLYSIS OF WATER AND AQUEOUS SOLUTIONS 103
9. RADIATION EFFECTS ON ORGANIC COMPOUNDS 111
10. EFFECTS OF RADIATION ON BIOLOGICAL POLYMERS 145
11. THE LIVING CELL 171
12. RADIATION EFFECTS ON SINGLE CELLS 202
13. RADIATION EFFECTS ON PLANTS 230
14. RADIATION EFFECTS ON ANIMALS 257

Part IV. Radiation Processing Technology

15. THE NATURE OF FOOD AND THE ROLE OF RADIATION 283
16. WHOLESOMENESS AND ACCEPTANCE OF IRRADIATED TREATED FOOD 300
17. RADIATION EFFECTS ON PACKAGING MATERIALS 311
18. FOOD IRRADIATION TECHNOLOGY 324
19. INDUSTRIAL RADIATION TECHNOLOGY 345
20. RADIATION FACILITIES AND PROCESS ECONOMICS 359
21. RADIOLOGICAL SAFETY 375
INDEX 395

List of Illustrations

Frontispiece. A nuclear reactor yields power and useful by-products of radiation and radioisotopes

FIGURE PAGE

1. Distribution of energy radiated by the sun 4
2. Atmospheric carbon-14 cycle as a consequence of nuclear emission 12
3. Fission yield curve for uranium-235 and slow neutrons . . . 14
4. Half-life curve for radioisotope 16
5. Atomic orbitals or wave functions plotted in three dimensional space 22
6. Schematic diagram of ionization 27
7. Geometry of molecules 30
8. Helical configuration of protein molecule 31
9. Molecular stretching and bending vibrations schematically shown in series of "movie stills" 33
10. Comparative strength of bonds in ethane 35
11. Distribution of energy of electrons ejected by collision process 47
12. Bremsstrahlung or deceleration radiation 48
13. Nuclear photo effect 52
14. Schematic diagram of tracks of ionizing particles 55
15. Sequence of events in thermal reactor core 63
16. Fuel assembly units of enriched uranium for reactors 65
17. Materials testing reactor, Arco, Idaho 66

18A. Schematic diagram of materials testing reactor 67

18B. Schematic diagram of materials testing reactor 68

19. A uranium-235 reactor 69
20. Schematic diagram of Van de Graaff electron accelerator . . 76
21. Linear accelerator, 50 Mev electrons 77
22. Dose-depth relation for electrons and gamma ray sources . . 78
23. Schematic diagram of resonant transformer 79
24. Schematic diagram of capacitron 80
25. Penetrating characteristics of electron beams 83
26. Dosage distribution by 3 Mev cathode (electron) and gamma rays in flour 84

27A. Dosimetry with cobalt glass strips 92
27B. Dosimetry with cobalt glass strips 93
28. Isodose curves for 6 × 7-inch cylinder in gamma field 94
29. Optimum distribution of electron energy across three-inch container with unscanned beam 95
30. Distribution of electron energy across 5-inch cylindrical container using scanning beam 96
31. Bombardment of container from two sides 97
32. Hydrogen peroxide yield altered by inhibiting back reaction, precipitating hydrogen peroxide as it is formed 104
33. Effect of iodine on H_2 yield from cyclohexane 114
34. Reactions of alkoxy radicals 118
35. Action of electrons on hexoses 128
36. Action of cathode rays on D-glucitol 128
37. Schematic diagram of radiation effect on hemoglobin 138
38. Radiation effects of pigments of red meat 139
39A. Effect of 1.5 megarads on iodometric properties of two per cent aqueous starch solution buffered at pH 4.7 with sodium acetate 148
39B. Rate of alpha amylase activity upon irradiated and control starch solutions 148
40. Schematic diagram of a protein molecule in various stages of denaturation 153
41. Per cent of amino acid survival on one per cent insulin as a function of irradiation dose 155
42. Radiation effects on fresh egg protein 156
43. Comparison of rennet coagulation times for irradiated and heated milk 158
44. A plot comparing the observed molecular weight for a wide variety of bombardment substances 163
45. Effect of temperature on the inactivation of 10 mg per ml solution of pepsin by 2,500,000 rads 164
46. Effects of irradiation on enzyme-substrate reaction rates (Enzyme trypsin; substrate-casein; dose–1.5 megarads) . . . 166
47. Schematic diagram of animal cell 172
48. Schematic diagram of plant cell 173
49. Relative complexity of organisms in terms of the number of atoms present 178
50. Schematic diagram of pathway of carbon compounds from carbon dioxide to glucose in photosynthesis 181
51. Pathway of carbon in respiration 182

52. Flow pattern of the Krebs cycle for cells growing on glucose as carbon and energy cycle 183
53. Metabolic pathway in plants 183
54. Phosphate bond energy harvested from respiration of sugar 184
55. Schematic representation of cell membrane surface 186
56. Protoplasm growth patterns 190
57. Schematic diagram of mitosis 192
58. Schematic representation of mutations 194
59. Radiation induced mutations 195
60. Schematic diagram of Crick-Watson double-threaded DNA . 196
61. Schematic diagram of duplication process for DNA particle. Crick-Watson model 197
62. Chromosomes sensitive to radiation effects 203
63. Radiation effect on cell when exposed to 350 rads at anaphase 203
64. Influence of presence of moisture on radiation resistance of bacterial spores 209
65. Relative radiation resistance of spores vs. vegetative cells of *Bacillus coagulans* 212
66. Influence of concentration in radiation resistance of *Bacillus subtilis* spores in neutral buffer solution 213
67. Effect of added biotin on growth of irradiated spores 215
68. Radiation destruction of bacterial spores—the D_M value—for *Clostridium botulinum* 219
69. Radiation destruction of fruit pathogens 222
70. Destruction of moist yeast cells with irradiation 223
71. Relation between virus diameter and dose required to inactivate 37 per cent of virus 224
72. Cobalt-60 irradiation field 231
73. Biological expression of absorbed high energy radiations . . 232
74. Changes in free auxin level immediately following X-irradiation of whole plant shoots or segments 234
75. Relative free auxin levels of plants following irradiation . . . 236
76. Possible loci of biochemical damage in the inhibition of plant growth by ionizing radiations 237
77. Growth of plant inversely proportional to the dose rate—above threshold value 238
78. Stem swellings and adventitious root production in *Xanthium* stems 239
79. Relation between the dose rate applied per day and the per cent tumorous plants after various periods of irradiation . . 240

80. Vegetative growth occurs in normal floral position following irradiation. When removed from gamma field, and allowed to recover, fused shoots rise in floral bud 241
81. Modified axillary bud expression following irradiation of *Trandescantia paludoso,* adventitious buds formed producing irregular leaves and multiple shoots (37 rads/day, for 90 days) 241
82. Floral abnormality in *Oxalis* due to irradiation of floral bud 242
83. Leaf abnormalities resulting from chronic radiation damage 243
84. Treated and control potatoes after one year's storage at 41°F. 244
85. Two year old Idaho Russet Burbank potatoes at 41°F., 85% R.H. 248
86. Relation of dose applied to fruits and their rates of respiration (tomato) 249
87. Respiration values for tomato fruits irradiated at two doses at zero time 250
88. Radiation pasteurization of strawberries decreases losses and extends distribution life 252
89. White blood cells response in humans exposed to single acute doses of radiation ranging from threshold to LD_{100} 263
90. Radiation effects on *Trichinella spiralis* 270
91. Insect disinfestation of beans with low level gamma irradiation 276
92. Comparative essential amino acid composition of human, beef, corn, and wheat proteins 286
93. Schematic diagram of test program for establishing wholesomeness and acceptance of irradiated foods for industrial application 302
94. Decrease in radiation flavor in beef during storage-temperature conditioning 304
95. Tenderness control in poultry meat 304
96. Radiation sterilized shrimp stored one year at room temperature (4.8 megarads) 305
97. Mold destruction in bread rolls 306
98. Radiation permits preservation of whole animal or large portions of animals (ham shown) 307
99. Improved refrigerated storage and distribution life of sliced meat (after two weeks) 308
100. Radiation permits room temperature storage of chickens . . 309
101. Electron sterilization of plastic containers 322
102. The D_M value for *Clostridium botulinum* 326
103. The D_E value for enzymes 327

104. Destruction of enzymes with moist heat. Ascorbic acid oxidase shown 329
105. Radiation sterilized intravenous fluid administrator set . . . 348
106. Commercially produced radiation sterilized tubing for medical applications 349
107. Molding of clothing can be eliminated with radiation treatment of packaged material 349
108. Early radiation facility using spent fuel elements at Idaho Falls, Idaho 360
109. High intensity gamma radiation facility at Dugway Proving Grounds, Dugway, Utah 361
110. Mobile irradiation processing plant 362
111. High specific activity radionucleide facilities will "feed" lower intensity facilities, improving costs of radiation processing 363
112. Spent fuel element radiation facility—interception of fuel elements between reactor and chemical reprocessing plant . . 364
113. Reactor core loop—a permanent radiation site 365
114. Mobile radiation source. Radionucleides or electron generators contained in mobile beds or vans. Shielding required as integral parts of the structures 365
115. Relation of cost to size in an irradiation facility 370
116 "Cutie pie" or gun type survey meter 378
117. Portal monitor—useful in detecting radioactive contamination on personnel as they pass through area 379
118. Beta-Gamma hand and foot monitor 381
119. Film badge—each unit contains two films 383
120. Pocket dosimeter and dosimeter charger 384
121. Alpha, beta, gamma survey meter 384
122. Shielding requirements for high energy electrons in several materials 385
123. Shielding requirements for gamma radiation to attenuate by a factor of ten 386

PART I

Introduction to Radiation and Its Effects

CHAPTER 1

Introduction

The Nature of Radiation

Man has always lived in a field of radiation and has been influenced by it. While he has but recently become aware of its presence, he is beginning to apply it to his own advantage.

What is radiation? In classical definition, radiation is the flow of energy from one point to another across empty space. Its flow requires no guidance as does the flow of water or the conduction of heat. Some radiations can flow through matter, virtually ignoring its existence. Witness the image of the landscape as seen through a clear glass window and compare it with that seen through an open door. Witness the shielding required around a nuclear reactor. The flow of radiant energy can occur as a traveling electromagnetic wave or as a traveling particle. The former type is represented by light waves, heat rays, radio signals and X-rays, all of which are qualitatively similar. The traveling particle type of energy flow includes a flying baseball, which contains much energy, as many broken windows have demonstrated. On the sub-microscopic scale, every moving particle also carries energy. Since radiation is defined as the organized flow of energy, it is necessary that the movement of the particles be organized so that their energy is in fact flowing from one point to another. This movement need not require a confining guidance.

Since the advent of the atomic age, the term radiation has been used in its narrowest definition, restricting it to those electromagnetic waves of higher energy, such as X-rays and gamma rays, and narrowing the class of particles to those of atomic size and smaller. It is radiation by this definition which is the subject of this book.

The World of Radiation

The sun, which is our primary energy source, delivers this energy as radiation. The earth receives this radiation primarily as electromagnetic waves ranging from radiowaves, through heat and light waves, out to very penetrating X-rays. Fortunately, the atmosphere above us shields mankind from the harmful X-rays; it also absorbs a considerable quantity of other wave lengths as well. The relative amounts of the various types of electromagnetic radiation from the sun which enter the earth's atmosphere are shown in Fig. 1. The amount of radiation received at the boundary of our world, the edge of the upper atmosphere, is two calories per sq. cm. per minute. This, when added up over the sunny side of our earth represents an energy contribution equivalent to 13 × 10^{23} calories per year, some of which is captured in photosynthesis and in the form of heat.

Cosmic Radiation

In addition to the wave radiation the sun also emits a very small but significant amount of particulate radiation. It is a component of so-called cosmic radiation. This radiation consists of streams of hydrogen nuclei, or protons, which are periodically hurled forth when an area of thousands of square miles of the sun's surface is convulsed in a magnetic storm, seen from the earth as a tiny dark spot. These streams of particles collide with the molecules of our atmosphere, causing some to break apart, transferring some energy to others which is then re-emitted as visible light. Since the protons are charged, they are guided by the earth's magnetic field towards the northern (and southern) regions, and there generate the well-known northern lights.

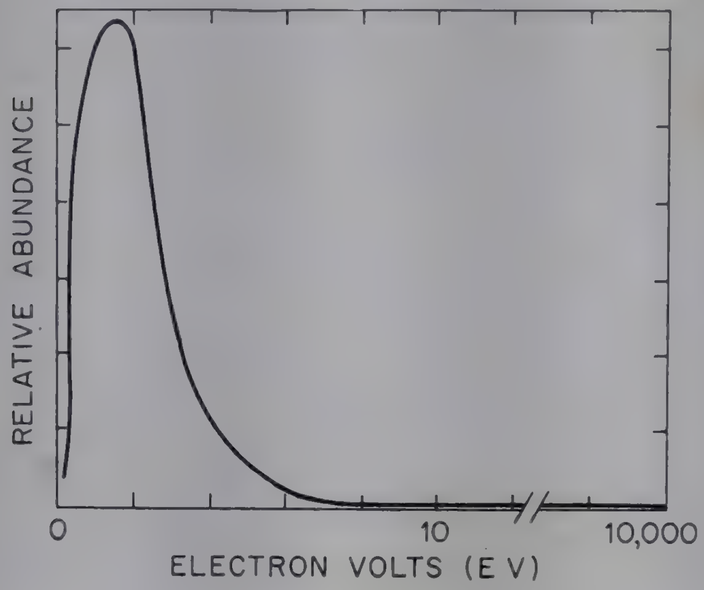


FIG. 1. DISTRIBUTION OF ENERGY RADIATED BY THE SUN

The earth is also being bombarded by a variety of heavier atomic particles originating in outer space. Many of these particles travel with more energy than can be generated by the most powerful particle accelerators yet built by man. On entering the atmosphere, these extremely energetic particles collide with air molecules and form so-called cosmic ray showers. These "showers" are formed when a single cosmic ray collides with a nucleus and forms several secondary particles; these in turn undergo further collisions, forming exponentially more particles, and so on.

Although the solar and stellar cosmic rays are not a significant energy source for the earth, they are nevertheless important in two aspects. First, the level of cosmic radiation is such that every human being is undergoing a relatively constant bombardment by this high energy radiation, receiving roughly fifty cosmic ray impacts during every second of his life. Further, one of the processes induced in the atmosphere by cosmic rays is the production of a radioactive isotope of carbon. This radioactive carbon is chemically transformed into carbon dioxide which is converted by plants into organic compounds which are then consumed as human food. Thus, an inevitable consequence of the life cycle is the incorporation of the radioactive carbon into the very substance of the body. Only when a human dies does he stop replenishing his radioactive carbon content and, subsequently, over hundreds and thousands of years, his remains become gradually less radioactive. It is this gradual "cooling off" after his death that makes possible the radiocarbon dating techniques which have contributed much to man's understanding of his ancestry.

Man also carries other types of radioactivity. A number of mineral constituents of the body, such as the teeth and the skeleton, contain elements, notably potassium, which occur naturally in slightly radioactive form (without the intervention of man-made nuclear explosions). Also, there are a number of radioactive elements present which, though not incorporated into the body, subject the individual to radiation. The amateur uranium prospector is receiving as much or more radiation than the hand-held Geiger counter which leads him unerringly to the richest deposit.

With the birth of the atomic age, there have become available massive sources of radiation of unprecedented intensity. Before World War II, a hospital was considered fortunate to have available a single gram of radium for therapy. The world supply at that time was several pounds. Today, most hospitals can have radiation sources with an intensity equalling that of the formerly available world's supply of radium. The widespread application of artificial radioactive materials in medicine, in science and industry is underway. Paralleling this vastly expanded production of artificial radio-elements there have become available commercially a variety of powerful radiation-generating machines or accelerators such as the linear accelerator, the Van de Graaff accelerator and the cyclotron.

Since the end of World War II there has been a considerable research effort directed towards understanding the effects of radiation on all forms of material, living and non-living. The results of these efforts have already yielded a number of irradiation processes employed in industry.

A somewhat larger number of processes are close to the point of finding industrial acceptance, and many more are in various stages of research and development. Those in the fields of food, agriculture, and biology and allied areas will be explored in this book.

Effects of Radiation

Radiation is unique in its ability to create highly reactive chemical species in matter through which it passes. The vast potentialities of radiation can be indicated by a listing of some of the effects that this agent can produce.

The physical properties of materials may be altered by radiation. Structural materials such as metals may be made brittle. Some plastics are made stronger and more durable, others lose mechanical strength and decompose. Certain solid materials suffer structural changes which can change their catalytic activity. Transparent substances such as glass and certain clear crystalline materials may become highly colored. Radiation can induce an electric current in certain semiconductor materials.

Numerous chemical changes can be produced by radiation. A mixture of nitrogen and oxygen, such as ordinary air, can be partially converted to nitrogen oxides which are important in the fertilizer industry. Ethylene gas can be converted to polyethylene plastic. Benzene and chlorine gas can be irradiated to yield the insecticide benzene hexachloride. Wood alcohol on irradiation is partially converted to ethylene glycol, an antifreeze.

The detailed biological effects of radiation are equally numerous. The effect which has received the most widespread recognition is the capability of killing living organisms, from single cells to large animals. When properly applied this capability is quite useful; a number of surgical supplies are being sterilized by irradiation, and the research on radiation preservation of foodstuffs has indicated many promising applications.

BIBLIOGRAPHY

ABETTI, G. 1957. The Sun. Second Ed. Macmillan Co., New York.
HOYLE, F. 1955. Frontiers of Astronomy. Harper and Brothers, New York.
LE PRINCE-RINGUET, L. 1958. Cosmic Rays. Prentice-Hall, Inc., New York.
SANDERSON, J. A., and HULBURT, E. O. 1955. Sunlight as a Source of Radiation. Radiation Biology. A. Hollaender, Editor, McGraw-Hill Book Co., Inc., New York.

CHAPTER 2

Nuclear Structure and Radioactivity

What Is Matter?

This question has been posed by many people in many places at many times. To a structural engineer an adequate answer might be that it consists of a variety of ponderable substances in a gaseous, liquid or solid condition displaying a wide variation in density and mechanical properties. A biologist may be satisfied to consider it as all substance divided broadly into two classes depending upon whether it displays animate or inanimate properties. A chemist finds it useful to view it as a seemingly infinite variety of combinations of one or more of some one hundred chemical elements. And a nuclear physicist may view the totality of matter as a set of combinations of a small number of so-called fundamental particles. Depending on the point of view, the motivation and the tools of observation, a variety of acceptable answers are possible. And a goal of scientific endeavor is to incorporate the various requirements of different points of view or, equivalently, techniques of observation into one consistent set of assumptions capable of reproducing all scientific experience. This program is, of course, far from completion.

A tentative set of answers may be given which is adequate for the purposes of this discussion. In connection with radiation and radioactivity we shall employ the fundamental particle picture. The discussion of physical and chemical effects of radiation will be phrased in the terminology of the atomic and molecular structure of matter. Lastly, the biological aspects of radiation will be treated in biological terms of the cell and the multicellular organism with emphasis on the metabolic pathways of living organisms where appropriate.

Matter, then, consists of very small particles called atoms, the smallest unit capable of displaying its chemical properties in bulk. The atoms, in turn, consist of an even smaller central portion called the nucleus, which contains almost all the mass of an atom, surrounded by a cloud of particles called electrons. Atoms combine in smaller or larger numbers to form molecules. The molecules are the smallest representatives of the thousands upon thousands of chemical substances common to our experience. In any one chemical substance all molecules are alike in that they all have the same number and spatial arrangement of various component atoms. The number of atoms in a single molecule may range from one atom to millions, the latter extreme being usually termed a macromolecule. Living organisms consist of specific complex aggregates of molecules including a variety of both ordinary small molecules such as water and oxygen and macromolecules such as proteins and carbohydrates.

Nuclear Structure

The atomic nucleus is composed of two kinds of particles, positively charged protons and uncharged neutrons. Electrical neutrality of atoms is maintained by the negatively charged electrons surrounding the nucleus. The masses of the three particles are:

Proton	1.6730×10^{-24} gm.
Neutron	1.6752×10^{-24} gm.
Electron	9.106×10^{-28} gm.

It is evident that the weight of an atom is concentrated in the nucleus.

Two numerical quantities are generally used to describe a nucleus, the atomic number Z, which is equal to the number of protons or positive charges in the nucleus, and the mass number M, which is the total number of particles (protons and neutrons) in the nucleus. The atomic number is also equal to the number of electrons surrounding the nucleus. Since the number of electrons determine the chemical properties of an atom, the atomic number may be regarded as a code number for the more familiar names of the chemical elements. Thus the element of atomic number 1 is hydrogen, that of atomic number 8 is oxygen, and that with an atomic number of 100 is Fermium, one of the most recent man-made chemical elements.

Many of the atomic nuclei can exist in a variety of forms called isotopes, which have the same atomic number (or number of protons) but a different number of neutrons. The isotopes of any element have different masses, but almost identical chemical properties. There are distinct but very small differences in such properties as vapor pressure, boiling point, and the rates at which the isotopic forms undergo chemical reactions. These slight differences in properties have been employed on a large scale to separate the pure isotopic species, notably the separation in nearly pure form of the uranium isotope of mass number 235 (92 protons and 143 neutrons) from its far more abundant companion isotope 238 (92 protons and 146 neutrons) by their slight difference in diffusion velocity in gaseous form.

The forces holding electrons in the vicinity of a nucleus are the familiar electrical forces, according to which unlike charges attract one another. On the other hand, the corollary that like charges repel one another clearly indicates that conventional electrical forces cannot account for the stability of the nucleus. It is the currently accepted view that the conventional electrical forces are the only forces that act between two particles separated no less than about 10^{-12} cm., roughly a nuclear diameter but that, and most importantly, at shorter distances in which two or more particles exist in the same nucleus new types of nuclear forces also come into play. If one considers now that the electrical energy of two particles varies inversely with the separation between them, and that the electrical forces do hold to distances of the order of a nuclear diameter, it is apparent that the binding forces in the nucleus are far greater than those between the nucleus and the surrounding electrons. A measure of energy commonly employed in nuclear, atomic and molecular energetics is the electron volt. The magnitude of the energies of various processes and structures of interest is indicated in Table 1.

TABLE 1

SELECTED NUCLEAR, ATOMIC AND MOLECULAR ENERGIES

Process or Structure	Approximate Energy in Electron Volts
Kinetic energy of a molecule at room temperature	0.02
Strength of a chemical bond	1–4
Activation energy of a chemical reaction	0–4
Energy of a photon of visible light	1–4
Binding energies of atomic and molecular outer electrons	5–20
Binding energies of atomic and molecular inner electrons	15–150,000
Kinetic energy of a particle at 1,000,000 °C.	130
Binding energy of deuterium nucleus	2,200,000
Energy of an emitted alpha particle	4,000,000–9,000,000
Energy of a gamma ray photon	500,000–5,000,000
Kinetic energy of a cosmic ray particle	20,000,000,000

The electron volt is defined as the energy acquired by an electron in falling through a potential of one volt. Since the electron has a very small mass (9.01×10^{-28} gm.) this unit is exceedingly small in magnitude.

$$\begin{aligned} 1 \text{ ev} &= 1.6 \times 10^{-12} \text{ ergs} \\ 1 \text{ erg} &= 6.25 \times 10^{11} \text{ ev} \end{aligned} \tag{1}$$

$$1 \, \frac{\text{erg}}{\text{sec.}} = 1 \times 10^{-7} \text{ watt} \tag{2}$$

$$\begin{aligned} 1 \text{ watt-sec.} &= 1 \times 10^{7} \text{ ergs} \\ 1 \text{ kwh.} &= 3.60 \times 10^{13} \text{ ergs} \\ 1 \text{ kwh.} &= 3.60 \times 6.25 \times 10^{24} \text{ ev} \\ 1 \text{ kwh.} &= 2.25 \times 10^{25} \text{ ev} \end{aligned}$$

Nuclear Structure and Binding Energies

Although there is no detailed theory available for the nature and origin of nuclear forces, a large body of experimental evidence has developed which exhibits many empirical regularities. One type of information is available from studies of nuclear binding energies. It was predicted by Einstein many years ago that mass and energy were interconvertible and related by the now famous equation

$$E = mc^2$$

where E is energy, m is mass, and c is the velocity of light. On the molecular scale of events, the energy changes occurring in various processes such as chemical reactions, are too small to lead to measurable mass differences. On the nuclear scale, however, the energy changes are large enough and the tools of measurement of sufficient precision to lead to accurately measurable changes and also to provide information on the magnitude of the binding energies characterizing various nuclear structures. For example, on the atomic mass scale (in which the oxygen atom is arbitrarily assigned the mass 16.00000) the mass of a proton is 1.00754 and the mass of a neutron is 1.00894. On the other hand, the mass of a deuteron which is a combination of a proton and a neutron is 2.0142. This deuteron mass is 0.00228 unit less than the combined masses of the proton and neutron. The decrease in mass is the result of energy liberated in the process of combination of these two fundamental particles. According to the Einstein equation this mass decrease is equal to an energy of about 2.2 million electron volts (Mev). That this is indeed the binding energy of the two particles has been independently verified by measuring the gamma ray energy required to break up the deuteron into a proton and a neutron.

By means of precise mass measurements the binding energies of various nuclei have been determined. It is found that starting with a single proton, a neutron can be added with the release of 2.2 Mev of energy. Further addition of either a proton or a neutron releases some further energy. But when a third particle is added, making four in all, an exceptionally strong structure is formed only if two of the particles are protons and two are neutrons. This four particle structure of atomic number two is an alpha particle and is also the nucleus of the helium atom. Above mass number four there are additional regularities in which nuclei containing the so-called "magic numbers" of either 50, 82, or 126 neutrons or protons or both exhibit unusually high stability or binding energies. These regularities of greater and lesser nuclear stability have been explained in terms of the nuclear shell model of

Goeppert-Mayer and Jensen. According to this model, the neutrons and protons are arranged in the nucleus in various energy levels or orbits with only a fixed number of each permitted per level. The shell model has been successfully applied to many of the electric and magnetic properties of nuclei and appears to account for the fact that only narrow ranges of proton-neutron ratios occur in nuclei of different atomic number.

As the number of particles in the nucleus increases, the short range nuclear forces can no longer act among all particles, whereas the electrostatic forces are still operative among all particles. The latter forces, being repulsive, decrease the binding energy available for an added proton and tend to decrease the overall stability of the nucleus. It is found in nature that all the heavy nuclei are radioactive. A further consequence of the energy level structure of the nucleus and its electrostatic forces is that nuclei are not necessarily spherical in shape. Recent work with scattering of high energy particles by nuclei has revealed a considerable variety of nuclear shapes ranging from spheres to ellipsoids to dumbbell shapes.

Radioactivity

The difference in stability of various nuclei suggests the possibility that a given type of nucleus may acquire a more stable form by emitting individual elementary particles or groups of such particles. This type of process does in fact occur and is the subject of study of many nuclear scientists, both chemists and physicists. Historically, this phenomenon—radioactivity—was discovered and studied by Becquerel and the Curies in the late nineteenth and early twentieth century, at a time when it was not even established that an atom had a distinct nuclear and extranuclear structure.

The elementary processes of radioactivity are many in number. The preceding discussion suggests processes such as emission of a proton, a neutron, or the stable combination of two protons and two neutrons—the alpha particle. Many such processes as well as a number of other types have been discovered and studied. The new types include emission of various combinations of gamma rays, electrons, positrons, and neutrinos, and also fission reactions.

When the particles in a given nucleus are in the lowest possible set of energy levels, the nucleus is stable with respect to its identity; it cannot undergo any spontaneous process except by losing particles. If, however, one or more of the nuclear particles is in a higher energy level, the nucleus is in an excited state, containing more energy than the minimum amount required for its existence as an entity with a fixed number of protons and neutrons. This energy can be released in the

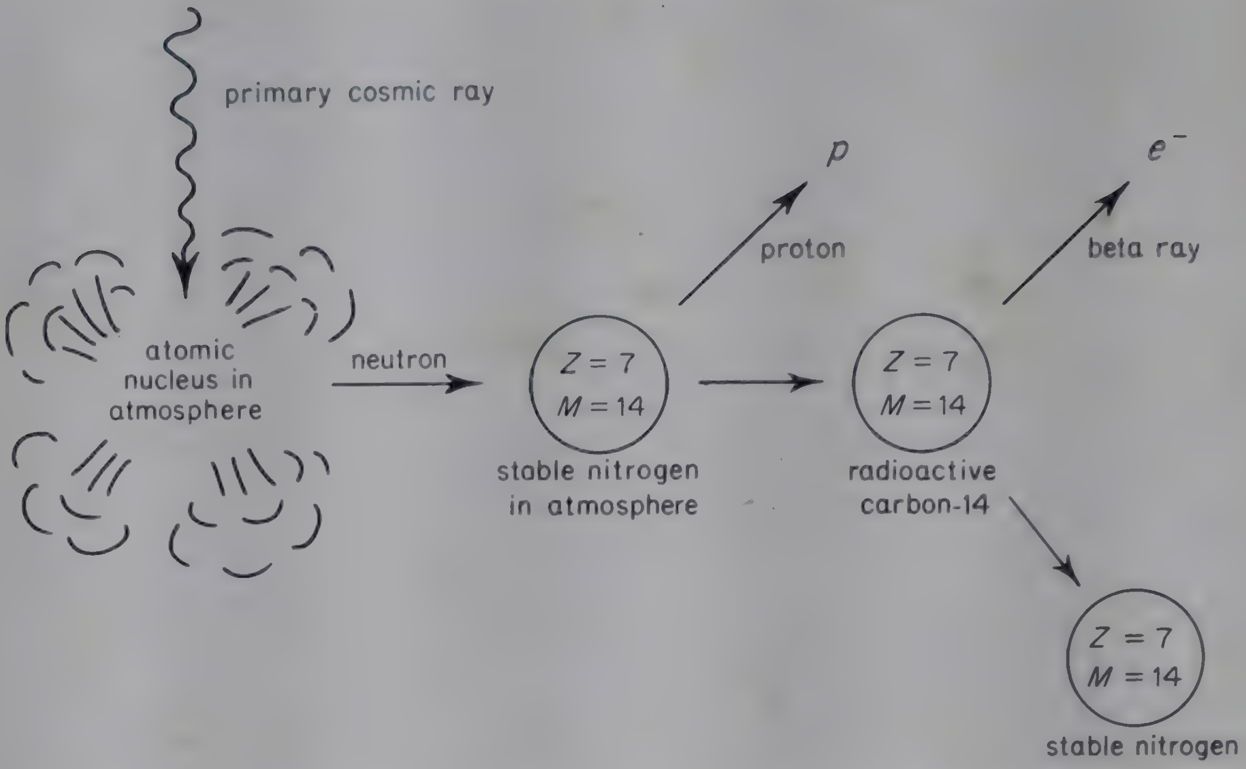


FIG. 2. ATMOSPHERIC CARBON-14 CYCLE AS A CONSEQUENCE OF NUCLEAR EMISSION

form of electromagnetic radiation called gamma radiation, the nucleus returning to the ground state in the process. This energy release is the nuclear analogue of emission of light, with only the difference that on the more grandiose nuclear energy scale this light contains much more energy and is, in consequence, of far shorter wave length than the visible light which is generated in an analogous process involving atomic or molecular electrons. An important point is that in neither case is the light stored as such in the excited nucleus or atom. The light or gamma radiation is "born" in the process of emission, and may be thought of as a bundle of energy emitted from the nucleus or atom with the velocity of light at the same time instant at which the nucleus assumes its ground state configuration (see Fig. 2).

It is a quite general consequence of modern physical theory that this form of energy emission occurs in distinct and definite amounts. When a number of particles occur in close association, as do the particles composing a nucleus or the electrons surrounding a given nucleus, the number of energy configurations these particles can assume is finite and the configurations grouped at definite energy intervals. Thus the amount of energy radiated in the process of transition from one energy level to a lower one is fixed in magnitude and assumes one or another particular value depending on the particular pair of energy levels involved. This principle is nothing more nor less than the principle of conservation of energy. The set of possible values of energy that can be radiated by a physical body such as a nucleus or an atom or a molecule is called its electromagnetic spectrum. Gamma emissions of various nuclei include examples where gamma photons of only one energy occur as well as cases where photons are emitted having any one of as many as ten different energies.

The electron has already been briefly described; it has a negative charge equal to that of the proton and a mass equal to only $^1/_{1,840}$ part of the proton. The positron has the same mass as the electron but a positive charge like the proton, and the neutrino has a mass probably equal to that of the electron or positron but has no charge. The discussion of nuclear structure was given in terms only of protons and neutrons. On the other hand, there is abundant evidence that these other particles are emitted by nuclei. Since there is reason to assume that they do not exist as such in the nucleus these particles must be assumed to be created at the instant of emission as was the case with gamma rays. The emission of a negative electron results in a unit increase in the positive nuclear charge Z, a positron emission produces a unit decrease in Z. In some cases, an electron and a positron may be emitted together from a nucleus resulting in no net change in atomic number. In those cases where only one or the other charged particle is emitted, it has been found necessary to assume for fundamental theoretical reasons, that an uncharged particle of similar mass, the neutrino, was simultaneously emitted. For many years the existence of the neutrino was based purely on circumstantial evidence and the belief that some of the very fundamental laws of physics were valid on the nuclear level. Very recent work appears to have produced direct experimental verification of the existence of this very elusive particle.

The natural occurrence of some radioactive isotopes and, in particular, the natural absence of some others has been a matter of interest for decades. With the discovery of artificially induced radioactivity it became evident that many more radioactive species could be produced in the laboratory than were found in nature. The reason for their absence also became clear and will be given below. Artificial radioactive isotopes are generally prepared by bombarding stable isotopes with various kinds of particles including neutrons, protons, deuterons, alpha particles and high energy gamma photons. In recent years considerable work has also been done on bombardment of isotopes with nuclei of heavier elements such as nitrogen and oxygen which consist of combinations of 14 and 16 particles, respectively. Except for neutrons and gamma rays, which are uncharged, the fact that both the bombarding particle and the target nucleus carry positive charges requires that large repulsive forces must be overcome in order to bring the projectile and target into close proximity. This necessitates accelerating these projectile particles to extremely high energies, of the order of several million electron volts. This acceleration is accomplished by any one of a variety of high energy accelerators such as a cyclotron, a linear accelerator or a synchrocyclotron.

Since neutrons carry no charge and hence are not subject to the electrostatic repulsive forces, they can interact with nuclei even though they may travel only with room temperature energy, of the order of several hundredths of an electron volt. Sources of neutrons consist primarily of nuclear reactors. The high energy requirement for gamma rays is set by the spacing between nuclear energy levels; these electromagnetic waves are comparatively unaffected by the charge of the target nucleus.

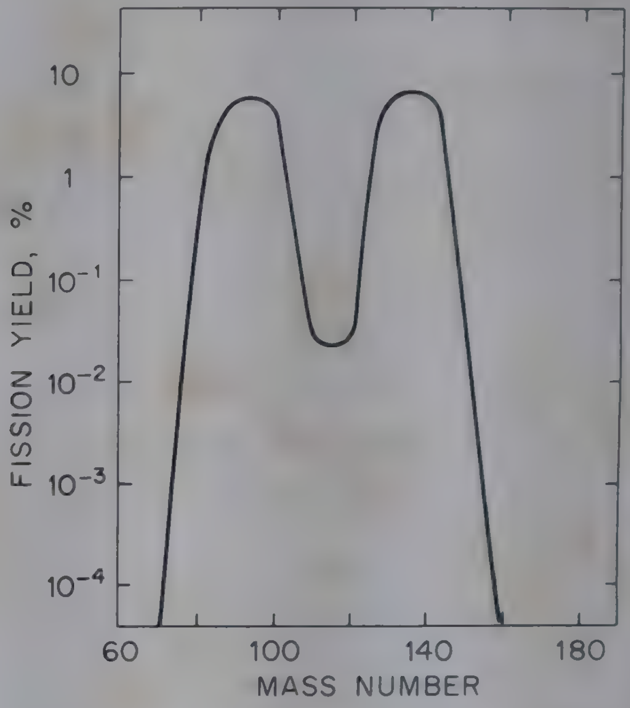


FIG. 3. FISSION YIELD CURVE FOR URANIUM-235 AND SLOW NEUTRONS

The processes occurring in the production of artificial radioactive isotopes are examples of nuclear reactions. The nuclear reactions are usually expressed in symbolic form by writing the incident and ejected particle in parentheses separated by a comma, leaving the corresponding mass number and atomic number changes implicit. For example, the reaction involving absorption of a neutron and subsequent emission of a gamma ray is written as

$$(n,\gamma)$$

and that involving absorption of an alpha particle followed by emission of a neutron is written as

$$(\alpha,n).$$

Examples of nuclear emissions and nuclear reactions, together with the corresponding atomic number and mass number changes are given in

Table 2. In the specific examples the chemical symbol is written instead of the atomic number, and the mass number is carried as a superscript.

Nuclear Fission

A type of nuclear reaction of great practical importance is fission. Heavy nuclei not only undergo small-particle reactions such as listed in Table 2, but can capture a neutron and split into two fragments of nearly equal size, liberating considerable energy in the process. The fact which makes a nuclear chain reaction possible is that in the fission process more neutrons are liberated which, in turn, can be captured by other nuclei and lead to more fission reaction. If the amount of material in which this fission process takes place is large enough so that the produced neutrons are captured by other nuclei before they can escape to the outside, a progressively more violent chain reaction may result. Without additional control a nuclear explosion occurs; however, with the use of suitable moderators and absorbers the reaction can be kept under control, yielding utilizable power in the form of heat.

A reactor converts uranium fuel into energy and a variety of fragments called fission products. The fission products consist of two distinct groups of nuclei having mass numbers near 95 and near 140 (see Fig. 3). This complex mixture of fission products is highly radioactive and, depending on the point of view taken, represents a serious radioactive hazard or a rich source of a variety of artificially produced radioisotopes. The increasing use of nuclear reactors has created a whole new technology of

TABLE 2

RADIOACTIVE DECAY AND NUCLEAR REACTION

Process	Mass Number Change	Atomic Number Change	Example
α-decay	-4	-2	$Po^{215} \rightarrow Pb^{211} + \alpha$
β-decay	0	$+1$	$H^3 \rightarrow He^3 + \beta^-$
γ-decay	0	0	$Co^{60} \rightarrow Co^{60} + \gamma$
positron decay	0	-1	$Ca^{39} \rightarrow K^{39} + \beta^+$
neutron decay	-1	0	$Br^{87} \rightarrow Br^{86} + n$
(n,α)	-3	-2	$Li^6\ (n,\alpha)\ H^3$
(n,γ)	$+1$	0	$Co^{59}\ (n,\gamma)\ Co^{60}$
(n,p)	0	-1	$N^{14}\ (n,p)\ C^{14}$
(α,n)	$+3$	$+2$	$F^{19}\ (\alpha,n)\ Na^{22}$
(γ,n)	-1	0	$H^2\ (\gamma,n)\ H^1$
(p,n)	0	-1	$Rb^{87}\ (p,n)\ Sr^{87}$
(d,n)	$+1$	$+1$	$C^{12}\ (d,n)\ N^{13}$
(d,p)	$+1$	0	$N^{15}\ (d,p)\ N^{16}$
(d,α)	-2	-1	$Mg^{24}\ (d,\alpha)\ Na^{22}$
$(d,2n)$	0	$+1$	$P^{31}\ (d,2n)\ S^{31}$
(α,p)	$+3$	$+1$	$A^{40}\ (\alpha,p)\ K^{43}$
$(\alpha,2n)$	$+2$	$+2$	$Cs^{133}\ (\alpha,2n)\ La^{135}$
fission	large	large	$U^{235} + (n, \text{fission})\ Cs^{137} + Rb^{97} + 2n$

reactor fuel element processing to recover "unburnt" uranium fuel and separate the radioactive fission fragments into usable form for tracer and radiation applications.

Half-lives

Radioactive isotopes decay at characteristic rates. Although for a single radioactive nucleus the decay process represents a unique event, never to be repeated, for large numbers of such nuclei a useful probabilistic concept can be applied. This concept is the half-life. The half-life *T* is the interval of time within which on the average one half of a given large number of such nuclei will undergo radioactive decomposition. This half-life is an intrinsic property of the particular type of radioisotope considered; it is independent of the state of chemical combination and of the temperature. Because of this independence of external conditions, the instantaneous rate of radioactive decay of a given mass of material depends only on the number of as yet undecomposed nuclei still present and their half-life T. This leads to an exponential law for the number N of undecomposed nuclei present at any time t later than some arbitrary starting time t_0 at which N_0 such nuclei are present.

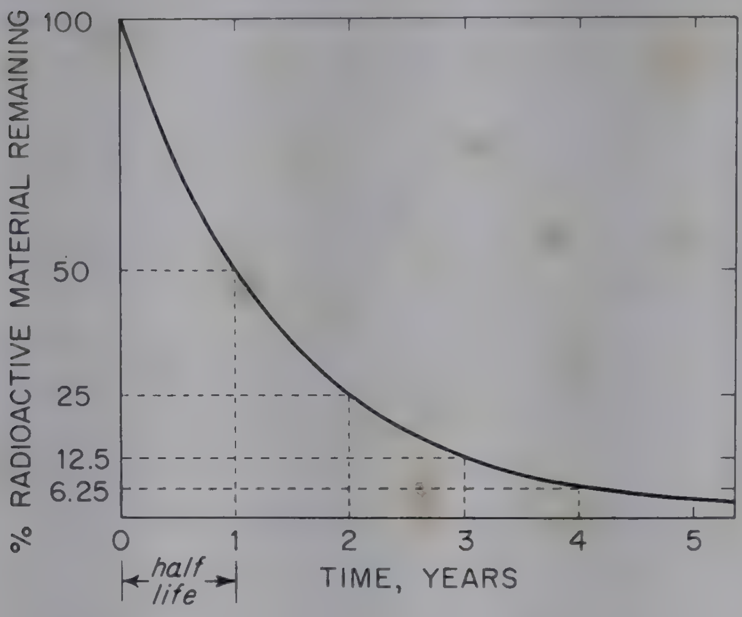


FIG. 4. HALF-LIFE CURVE FOR RADIOISOTOPE

$$N = N_0e^{-0.69315t/T}$$

An illustrative graph of this relation is given in Fig. 4 for a million radioactive nuclei decaying with a half-life of one year. The half-life of a radioisotope is a quantity of considerable technical importance. Since the intensity of radiation from a radioisotope depends on the number of still active nuclei present it is evident that the intensity also decreases exponentially. Further, the radiation intensity decreases more rapidly for radioisotopes with short half-lives than for those with long half-lives. On the other hand, the number of active nuclei required to produce a given instantaneous intensity of radiation will increase with the half-life of the isotope since a given number of nuclei of a longer lived isotope undergo fewer disintegrations in a given small time period than the same number of nuclei of a shorter lived isotope. The intrinsic properties of radioisotopes pose practical problems in technical applications. Isotopes are found to have half-lives ranging from a millionth of a second to millions of years. However, the conflicting considerations of intensity and long term stability reduces the useful number to rather few. The properties of some of the principal radioisotopes whose importance will be discussed later are given in Table 3.

Returning briefly to the problem of the absence of many radioisotopes in nature, the wide range of half-lives for various isotopes provides the clue. Current theories of the earth's origin indicate that the earth itself is about two billion years old. Clearly, even if all the now known radioisotopes were present at the time the earth was formed, only those having long half-lives would have survived in any detectable amounts. This appears to be a satisfactory explanation except for the C^{14} radioisotope. This isotope, as indicated earlier, is one of the few which are constantly being regenerated by cosmic ray bombardment of atmospheric nitrogen. The half-life of C^{14}, about 5,600 years, is short enough that, whatever the amount of this isotope present at the time of the origin of the earth, this starting quantity is essentially absent. The radioactive carbon cycle is in a relatively steady state with the principal source of replenishment due to steady cosmic ray bombardment. Since the atmos-

TABLE 3

PROPERTIES OF RADIOISOTOPES[1]

Isotope	Radiation (Energy in Mev)	Half Life
H^3	$0.0179\ \beta^-$	12 yr.
C^{14}	$0.156\ \beta^-$	5,600 yr.
P^{32}	$1.707\ \beta^-$	14.3 d.
S^{35}	$0.169\ \beta^-$	87 d.
K^{40}	$1.4\ \beta^-$	1.3×10^9 yr.
Ca^{45}	$0.260\ \beta^-$	160 d.
Co^{60}	$0.312\ \beta^-, 1.17\ \gamma, 1.33\ \gamma$	5.2 yr.
Sr^{90}	$0.61\ \beta^-$	28 yr.
Zr^{95}	$0.360\ \beta^-, 0.396\ \beta^-, 0.72\ \gamma, 0.76\ \gamma$	65 d.
Nb^{95}	$0.15\ \beta^-, 0.75\ \gamma, 0.92\ \gamma$	35 d.
I^{131}	$0.335\ \beta^-, 0.608\ \beta^-, 0.363\ \gamma, 0.638\ \gamma, 0.283\ \gamma$	8.0 d.
Cs^{137}	$0.52\ \beta^-$	30 yr.

[1] Adapted from Strominger, Hollander, and Seaborg (1958).

pheric carbon goes through its terrestrial cycle primarily as carbon dioxide, only those processes using atmospheric carbon dioxide would include this radioactive carbon. The two principal types of such processes include formation of limestone and other mineral carbonates and the photosynthetic cycle. Only the latter process involves a continual interchange of carbon dioxide with incorporated carbon so that the radioactive carbon cycle is in equilibrium with the atmospheric C^{14}. At death, this interchange ceases and the incorporated carbon decays without replenishment. This fact is the basis for the now famous radiocarbon dating method of Libby and the fascinating information it has yielded about our past, for example, the date agriculture was probably discovered.

Unit of Radioactive Intensity

The unit of radioactive intensity is the curie. This quantity was originally defined as the number of disintegrations per second which occur in one gram of pure radium. This has been adopted in the form of a more general definition:

$$1 \text{ curie} = 3.7 \times 10^{10} \text{ disintegrations per second}$$

This quantity is not adequate for radiation work since the rate of energy release depends not only on the rate of disintegration, but also on the energy liberated per disintegration, which is different for different materials. Nevertheless, the curie is a useful starting point from which to consider problems of energy release and other expressions of radiation intensity.

The units of radiation and methods of evaluation will be discussed in detail in Part II.

BIBLIOGRAPHY

BETHE, H., and MORRISON, P. M. 1956. Elementary Nuclear Theory. Second Ed. John Wiley and Sons, Inc., New York.

BLATT, J. M., and WEISSKOPF, V. 1955. Theoretical Nuclear Physics. John Wiley and Sons, Inc., New York.

EVANS, R. D. 1955. The Atomic Nucleus. McGraw-Hill Book Co., Inc., New York.

FRIEDLANDER, G., and KENNEDY, J. W. 1949. Introduction to Radiochemistry. John Wiley and Sons, Inc., New York.

GLASSTONE, S. 1958. Sourcebook on Atomic Energy. Second Ed. Van Nostrand, New York.

POLLARD, E. C., and DAVIDSON, W. L. 1951. Applied Nuclear Physics. Second Ed. John Wiley and Sons, Inc., New York.

RICE, F. O., and TELLER, E. 1949. The Structure of Matter. John Wiley and Sons, Inc., New York.

RICHTMYER, F. K., and KENNARD, E. H. 1947. Introduction to Modern Physics. Fourth Ed. McGraw-Hill Book Co., Inc., New York.

RUTHERFORD, E., CHADWICK, J., and ELLIS, C. 1930. Radiations from Radioactive Substances. The University Press, Macmillan, New York.

STROMINGER, D., HOLLANDER, J. M., and SEABORG, G. T. 1958. Table of Isotopes. Rev. Mod. Phys. *30*, 585–904.

CHAPTER 3

Atoms and Molecules

The arrangement of the electrons around the nucleus and the detailed behavior of this electronic arrangement determines many of the chemical and physical properties of matter. The development of the concept by Rutherford of the nuclear atom—a nucleus surrounded by electrons—provided the key to a satisfactory understanding of these properties. However, a revolutionary change in the basic concepts of theoretical physics was required before this understanding could be developed in quantitative form. This revolution—the development of quantum mechanics by Bohr and Sommerfeld—allowed the evolution of a concise theoretical framework which permitted for the first time the exact theoretical calculation of many properties of matter. It has been stated that insofar as quantum mechanics is correct, chemical questions are problems in applied mathematics.

The necessity for the revolution in physical theory required to describe adequately the properties of atoms may be indicated by the following consideration. A stationary negative electron could not remain separate from a positive nucleus, as the existing electrical forces would pull them together. If it is assumed that one electron can remain outside the nucleus by constantly traveling around it at high speed, another difficulty arises. According to classical theory, this orbiting electron would lose energy by radiating light, traveling in an ever-tightening spiral path, and eventually falling into the nucleus. The impasse between physical fact and classical theory was resolved by Bohr, who in 1913 founded a new atomic mechanics on the following two postulates:

1. An atomic system is stable only in a certain set of states, the "stationary states," which in general correspond to a discrete sequence of values of the energy of the atom. Every change in this energy is associated with a complete "transition" of the atom from one stationary state to the other.

2. The power of the atom to absorb and emit radiation is governed by the law that the radiation associated with a transition must be monochromatic and of frequency ν such that

$$h\nu = E_2 - E_1$$

where h is Planck's constant and E_1 and E_2 are the energies of the stationary states concerned.

It is seen that the first of these postulates accepts as a definition the very stability of atomic systems which could not be accounted for by classical theory. The second postulate relates the radiation absorbed and emitted by atoms to the energy content of a quantum of light, which was previously developed by Planck in connection with a study of the equilibrium between radiation and the matter which absorbs and emits it. His study was the first work in theoretical physics which required the concept of a quantum to successfully explain experimental findings. It was the first step in the departure from classical theory.

The atomic quantum initiated by Bohr in 1913 brilliantly accounted for the optical properties of the hydrogen atom and gave a clear explanation for many of the simple but mystifying regularities which many experimenters had discovered in the visible, ultra-violet and X-ray spectra of a number of elements. A further profound success of the Bohr theory was that, while on the atomic scale its quantum conditions amounted to a radical departure from classical physics (and agreed with experiment), the theory, when applied to large scale systems, gave the same results as classical theory. And classical theory, after all, successfully described whole areas of physical phenomena. However, it gradually became apparent that the Bohr theory, in which the hydrogen atom was treated like a miniature planetary system subject to the quantum conditions, could not at all account for the properties of atoms which bore more than one electron. Many attempts were made to develop the atomic-scale celestial mechanics of nuclei surrounded by more than one electron in such a way as to lead to answers without continually introducing new arbitrary assumptions.

Finally, in the middle 1920's, the dilemma was resolved in drastic fashion by a number of investigators—de Broglie, Schroedinger, Born, Jordan, Heisenberg, and Dirac, who, singly, or in small groups, developed a number of theories which constitute present-day quantum mechanics. The various theories were soon shown to differ more in form than in content. All contained the two postulates of Bohr.

The de Broglie-Schroedinger theory was based on an assumed wave-particle duality of all matter. De Broglie had suggested that all particles were accompanied by waves, and that in some sets of observations the particle aspect of matter predominated, and in others, the wave aspect. This was soon verified by Davisson and Germer who showed that electrons, hitherto considered purely particles, could under certain circumstances be diffracted and display interference phenomena of a purely wave nature. Schroedinger developed an equation for these de Broglie waves in which the Bohr stationary states and quantum numbers appeared automatically as the result of general reasonable physical and mathematical requirements.

According to the wave mechanical theory of Schroedinger, the various possible states of an electron are described by wave functions which are characterized by a set of values of three different quantum numbers. These three quantum numbers, called the principal, orbital and magnetic quantum numbers, can take on only integral values and only a restricted set of integral values of the latter two are allowed for a given value of the principal quantum number. The principal quantum number is related to the distance from the nucleus at which the electron spends most of its time. The orbital quantum number is related to the quantization of the orbital angular momentum of the electron as it travels around the nucleus in one of the stationary states. And the magnetic quantum number is related to the quantization of the angle with which the orbital plane can incline to an external magnetic field. An important departure from the Bohr picture is that the atom is no longer a microscopic planetary system; the electrons do not travel around the nucleus in precisely defined orbits; and the wave functions describe the electrons in probabilistic terms. Whereas in the Bohr theory one could consider the electron as traveling around the nucleus in a circular orbit of a particular radius r, the analogous wave mechanical description is that of a spherically symmetrical probability density (or wave function) in which the region of maximum probability occurs in the neighborhood of the radius r. Corresponding to the elliptical orbits of the Bohr theory, there are wave functions of lower symmetry (see Fig. 5).

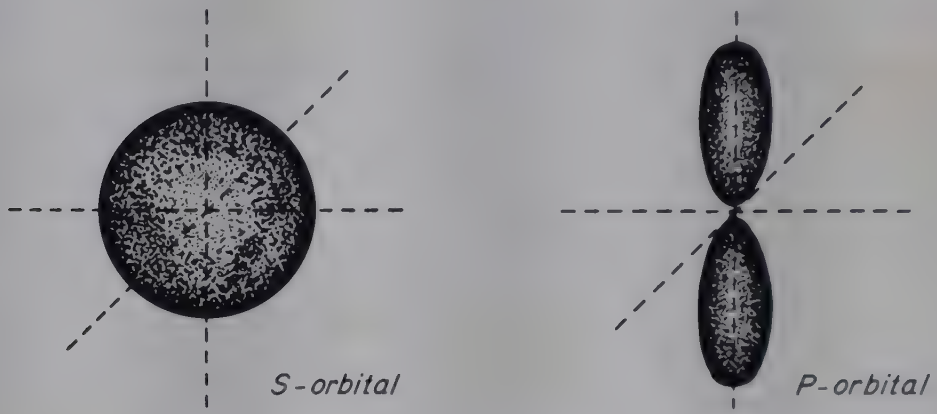


FIG. 5. ATOMIC ORBITALS OR WAVE FUNCTIONS PLOTTED IN THREE DIMENSIONAL SPACE

Probability of location of electron at any point is proportional to the darkness of the cloud shown schematically above.

The wave mechanical theory of the atom as developed by Schroedinger was still incomplete. The number of spectroscopic lines emitted by an atom in the presence of magnetic fields was greater than allowed by the number of stationary states corresponding to those of three quantum numbers. This discrepancy was resolved quantitatively by the suggestion of Uhlenbeck and Goudsmit that the electron itself behaved like a spinning charge which had a magnetic moment that was restricted (quantized) to two values. The spin was shown theoretically to be an intrinsic property of the electron when Dirac formulated his extension of quantum mechanics which incorporated the theory of relativity.

In the fully developed theory, four quantum numbers are required to describe the stationary state of an electron in an atom. These are:

n —the principal quantum number
l —the orbital angular momentum quantum number
m—the magnetic quantum number
s —the spin quantum number

The rules governing the sets of allowed values of these quantum numbers are:

n is an integer 1,2,3. . . .
l can assume integral values from 0 to $n - 1$
m can assume all integral values from $-l$ to $+l$
s is either $+{}^1/_2$ or $-{}^1/_2$

For example, for $n = 1$, l can only be 0, m must be zero, and s can be either $+{}^1/_2$ or $-{}^1/_2$. Electron states in an atom then can be concisely described by merely listing the values of the four quantum numbers in the form (n, l, m, s). For $n = 1$, the states are $(1, 0, 0, +{}^1/_2)$ and $(1, 0, 0, -{}^1/_2)$. For $n = 2$ there are eight states as follows:

$(2, 0, 0, +{}^1/_2)$ $(2, 1, 0, +{}^1/_2)$
$(2, 0, 0, -{}^1/_2)$ $(2, 1, 0, -{}^1/_2)$
$(2, 1, 1, +{}^1/_2)$ $(2, 1, -1, +{}^1/_2)$
$(2, 1, 1, -{}^1/_2)$ $(2, 1, -1, -{}^1/_2)$

One problem was yet to be solved. The theory so far gave an enumeration of the stationary states, analytical forms for the wave functions and methods for calculating the properties of a single electron in any of these states, but in the case of many-electron atoms, there remained the question of how the electrons were disposed among these states. Pauli gave the answer in his famous exclusion principle, according to which no two electrons could be in the same stationary state. With this principle there was a complete theoretical structure, which, from a concise set of postulates, was able to account for the regularities in the chemical and physical properties of all the chemical elements.

The Periodic Table

In the nineteenth century, a number of chemists had observed regularities in the chemical properties of the various elements. These could be grouped into classes such as the very reactive alkali metals (lithium, sodium, potassium, rubidium, cesium), the inert gases (helium, neon, argon, krypton, xenon), and the corrosive halogens (fluorine, chlorine, bromine, iodine). The most thorough presentation of these regularities was developed by Mendeleyev who, in 1861, arranged these elements in order of increasing atomic weight and found that with but very few exceptions, the repetition of chemical properties occurred in cycles of 8 or 18 elements. He assumed that this near-regularity was a law of nature and, arranging the known elements into a periodic table, predicted the existence of yet undiscovered elements which when inserted in particular places of the table would make the near-regularity a perfect one. On the basis of the assumed perfect regularity, he predicted the approximate atomic weight and the chemical properties of a number of elements. His predictions were verified within fifteen years by the discovery of three of the "missing" elements, gallium, scandium, and germanium. As recently as 1940 elements of atomic numbers 43 and 87 were still missing.

With the development of techniques for transmuting elements by neutron bombardment, the missing elements were artificially produced and their extreme radioactivity indicated the reason for their absence in nature. Their chemical properties, however, fitted perfectly into the scheme of Mendeleyev.

The development of atomic theory led very quickly to the insight that the relevant factor was not the atomic weight as once thought, but the atomic number of the element. The striking success of the Mendeleyev correlation was due to the accidental parallel between increasing atomic weight and atomic number. This new insight removed the few discrepancies in the periodic table where two adjoining elements were in reversed positions if ordered strictly according to their atomic weights.

One of the triumphs of the quantum mechanical theory of the atom is that it provides a fundamental explanation for the periodicity of chemical properties. The recurrence of similar properties at intervals of 2, 8, and 18 places in the table corresponded exactly to the number of electrons which could have the same principal quantum number. Not only the periodic recurrence of properties was accounted for, but also changes in properties from one element to the next became explainable.

This theory accounted not only for the gross structure of the periodic table but provided explanations for many of the detailed variations in the properties of the elements. A useful illustration is given by the variation of the first ionization potential of the elements, which is the energy necessary to remove the most loosely bound electron from an atom of the element. The first ionization potentials of the first 17 elements are given in Table 4.

TABLE 4

IONIZATION POTENTIALS OF FIRST 17 ELEMENTS IN PERIODIC SYSTEM[1]

Element	ev	Element	ev
H	13.6	Ne	21.6
He	24.6	Na	5.1
Li	5.4	Mg	7.6
Be	9.3	Al	6.0
B	8.3	Si	8.1
C	11.3	P	10.9
N	14.5	S	10.3
O	13.6	Cl	13.0
F	17.4		

[1] From American Institute of Physics Handbook Sect. 7. D. E. Gray, Ed. McGraw-Hill, 1957.

It is seen that the second element has a much higher ionization potential than the first and that the value of the third is much lower than the first two. As one proceeds from the third to the tenth element the values once more rise and at the eleventh element the ionization potential once more drops abruptly. The explanation for this variation is as follows. The first two elements can accommodate their electrons, one and two, respectively, in the orbital of principal quantum number one. In the case of helium, however, one electron cannot completely screen the unit increase in nuclear charge from the other electron, so both electrons see a greater nuclear charge than does the single hydrogen electron. They are thus more tightly bound. In the case of lithium, however, the additional electron must be accommodated in the orbital of principal quantum number two, in which the added electron is further removed from the nucleus. This sudden increase in distance, together with the fact that this larger distance makes the screening of the nucleus by the other two electrons more effective, results in a much lower ionization potential. As one proceeds through element number ten, neon, all these elements can add the required number of electrons into the second orbital and none of the added electrons can completely screen the parallel increase in nuclear charge from the other electrons in this orbital. The result is that in each successive element of this group the electrons see an increasing nuclear charge and are more tightly bound. With sodium, the additional electron must be accommodated in the orbital of quantum number three, resulting once more in a sudden increase in the distance from the nucleus and an increased effectiveness of screening. It would follow from this explanation that the trends in the binding of the electrons should also result in parallel trends of distances from the nucleus at which the outermost electrons are added to a principal orbital, the increased effective nuclear charge seen by each electron draws all of them closer to the nucleus (see Table 5).

TABLE 5

ATOMIC RADII FOR FIRST ELEVEN ELEMENTS[1]

Element	Covalent Radius	Metallic Radius
H	0.30×10^{-8} cm.	
He		rare gas radii not comparable
Li		1.58
Be		1.12
B	0.88	
C	0.77	
N	0.70	
O	0.66	
F	0.64	
Ne		rare gas radii not comparable
Na		1.92

[1] From Wells, A. F. (1947).

A similar, but reversed, set of trends exists in another quantity, the electron affinity. This is the ease with which an additional electron may be added to a given atom, making it a negative ion. Hydrogen has a slight positive electron affinity (it could barely hold on to an extra electron). Helium has none whatever. From lithium through nitrogen, the theoretically calculated values indicate very little, if any, affinity. Oxygen and fluorine, however, display affinities of nearly 3 and 4 electron volts. The affinity of neon has been calculated to be negative. The general nature of these trends can be qualitatively understood in terms

TABLE 6

ELECTRON AFFINITIES OF THE ELEMENTS[1]

Element	Electron Affinities, ev	Element	Electron Affinities, ev
H	0.73	Ne	−1.24
He	−0.53	Na	−0.08
Li	0.38	Mg	−0.91
Be	−0.81	Al	0.50
B	−0.09	Si	2.70
C	0.83	P	0.57
N	−0.10	S	2.17
O	0.70	Cl	3.05
F	2.93		

[1] From Pritchard, H. O. (1953).

of the same considerations of average distance of the nuclear charge by the other electrons (see Table 6).

Valence and Molecular Bonding

The concept of the molecule arose out of many sets of observations which indicated that different atoms tended to combine in the ratios of small numbers, to form chemical compounds. This concept, the existence of the molecule, led rapidly to the systematization and simplification of the problem of explaining the existence of thousands upon thousands of substances which had different chemical and physical properties. At the same time there arose the problem of understanding the nature of the forces which held these atomic groupings together. An early and reasonable suggestion was that these forces were electrostatic in nature. This was supported by observations that certain substances such as salt and hydrogen chloride could be dissolved in water, forming solutions which could conduct electricity.

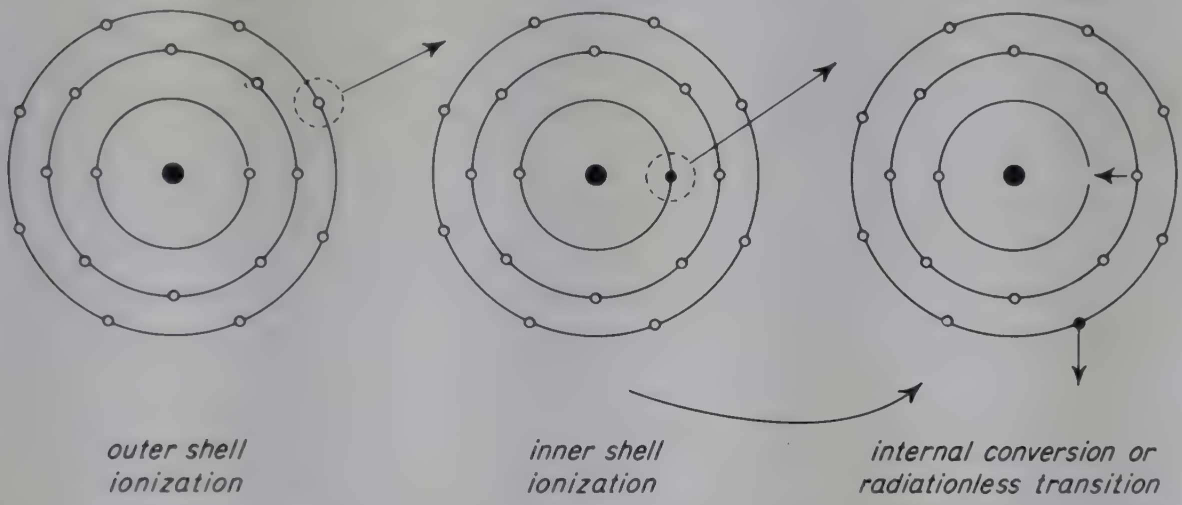


Fig. 6. Schematic Diagram of Ionization

However, the same observations which verified this hypothesis in the case of salt and hydrochloric acid demonstrated that other forces were involved in the much larger class of substances which did not ionize in solution (Fig. 6). The inadequacy of this hypotheses is clearly illustrated by examining the preceding information on ionization potentials and electron affinities. If an atom of low ionization potential such as lithium were brought into proximity with an atom of high electron affinity, such as fluorine, an electron could be spontaneously transferred from the lithium to the fluorine with the liberation of heat. The atoms would then be transformed into a positive and negative ion, respectively, which then would attract one another and form stable molecules. This is indeed so for these classes of compounds; they form substances which conform to the observations mentioned above. This approach, however, breaks down completely for groups of atoms which have disparate ionization potentials and electron affinities. The cases which most clearly violate this hypothesis are those of the elements themselves. Many of the elements, including all of the first eleven except helium and neon form stable molecules consisting of two or more atoms. In some cases, such as sulfur and germanium, stable molecules containing as many as eight atoms have been detected.

Throughout the nineteenth century, while the explanation of this problem remained hidden, chemists developed a set of valence numbers. These valence numbers, assigned to each element, were an index of the mysterious forces which tended to be saturated by combination with other atoms. An atom of valence four could combine with one other of valence four, or two of valence two, or one each of valence one and three, and so on. With only the slightly discomforting fact that some elements required more than one value of the valence number, this empirical concept for the first time really systematized the truly vast amount of experimental information on the composition of chemical compounds.

With the interpretation of the periodic table in terms of the electronic structure of atoms a major step in the solution of this problem was made by G. N. Lewis, who postulated that the chemical "bonds" implied by the valence numbers consisted each of a pair of electrons shared by two atoms. The atoms could each donate one electron to the pair, forming a covalent bond, or one atom could donate both electrons, forming a coordinate covalent bond. This postulate was rapidly developed into the electronic theory of valence by Lewis, Kossel, Langmuir, Sidgwick, and other chemists. The important observation was soon made that atoms tended to obtain "loan" of additional electrons by this sharing process in a number just sufficient to form an outermost electron shell containing eight electrons, owned or borrowed. The sole exception was hydrogen, for which this optimum number was two. The inert gases, helium, neon, argon, krypton, and xenon had this optimum number of electrons as atoms and thus would display no tendency to form compounds.

The now restricted problem of the stability of the electron pair bond was finally solved by use of quantum mechanics.

The application of quantum mechanics to the problems of molecular structure and stability led to the following results: The distribution of electrons among the various nuclei of the molecule could be described by probability distributions, or molecular orbitals, in a manner analogous to that applying to atoms. The electrons composing the inner shells of a particular atom remain distributed about the nucleus of the particular atom much in the same way as is the case in the isolated atom. Some of the outermost orbitals of the atoms combine to form molecular orbitals in which the electrons have a high probability of being located between the nuclei of any particular pair of atoms. The remaining outermost orbitals now form probability distributions which are directed away from adjacent atoms. The quantum numbers characterizing the atomic orbitals become transformed into molecular quantum numbers which, except for the spin, have a less direct physical interpretation than in the atomic case. However, the important result is obtained that stable chemical bonding generally occurs if two electrons occupy the orbital lying between atoms. And the pair of electrons occupying this orbital must have opposite spins. If these electrons have equal spins, they must occupy another orbital which is of such high energy that the adjacent atoms tend to repel one another. The orbitals lying between the nuclei are termed sigma orbitals. In this way, the electron pair bond of G. N. Lewis, was put on a firm theoretical basis.

The empirically observed tendency to complete an outermost shell of eight electrons, was justified semi-quantitatively by quantum mechanics on the basis of the amount of stabilization energy that would occur from the resulting molecular electron configuration. It was shown that the orbitals directed away from adjacent nuclei could interact with one another to form additional bonds between atoms, adding to the stability of the molecule. This, however, would occur only if, classically speaking, the resultant sharing of electrons would result in an eight electron outer shell for each atom.

Geometry of Molecules

The spatial arrangement of atoms composing a molecule is a matter of both intrinsic interest and considerable biological importance. In the case of diatomic molecules, there is of course no problem. In larger molecules however, the experimental results on molecular configurations appear sometimes to be in accord with esthetic considerations of symmetry, and frequently appear to violate them. The molecule carbon dioxide, CO_2, is a linear molecule consisting of a central carbon atom and two neighboring oxygens. The well-known molecule H_2O, however, is more v-shaped; the angle between hydrogens being about one hundred and five degrees. And the ammonia molecule, NH_3, has the configuration of a pyramid with the nitrogen atom at the apex, rather than a plane triangle of hydrogens with a nitrogen atom in the center (Fig. 7). This type of dissymmetry largely disappears when taking account of the fact that the location of the nuclei is dictated by the distribution of the molecular electrons. The oxygen atom has six electrons and forms a water molecule by

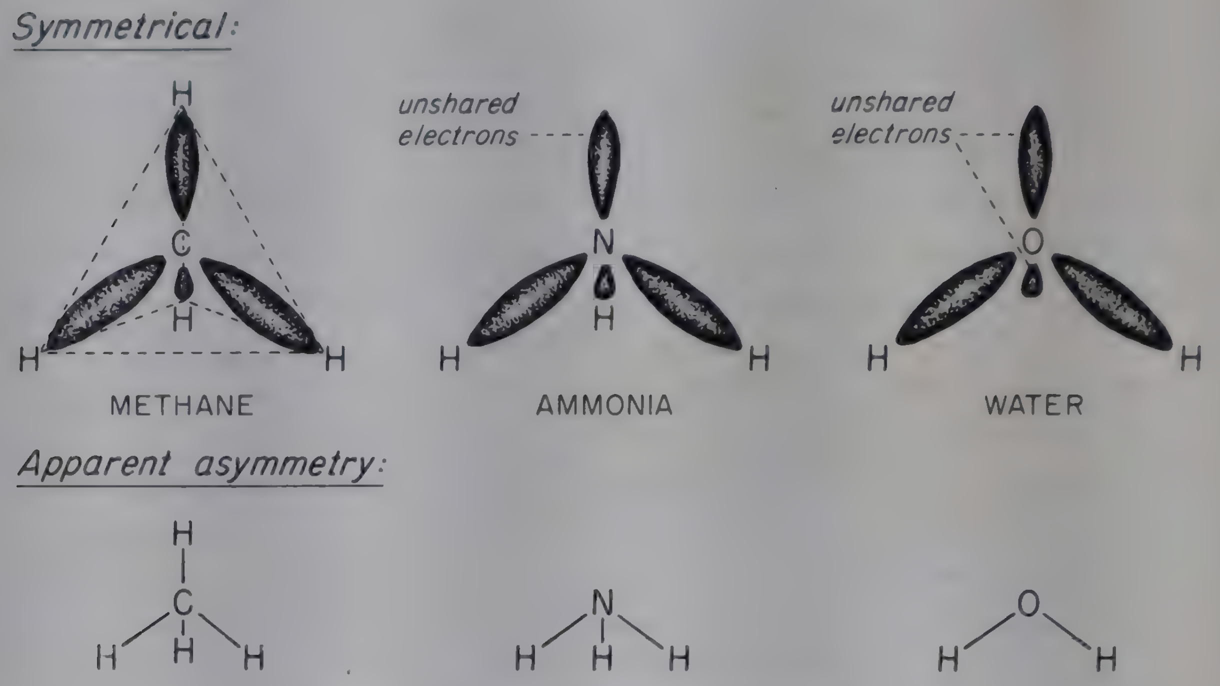


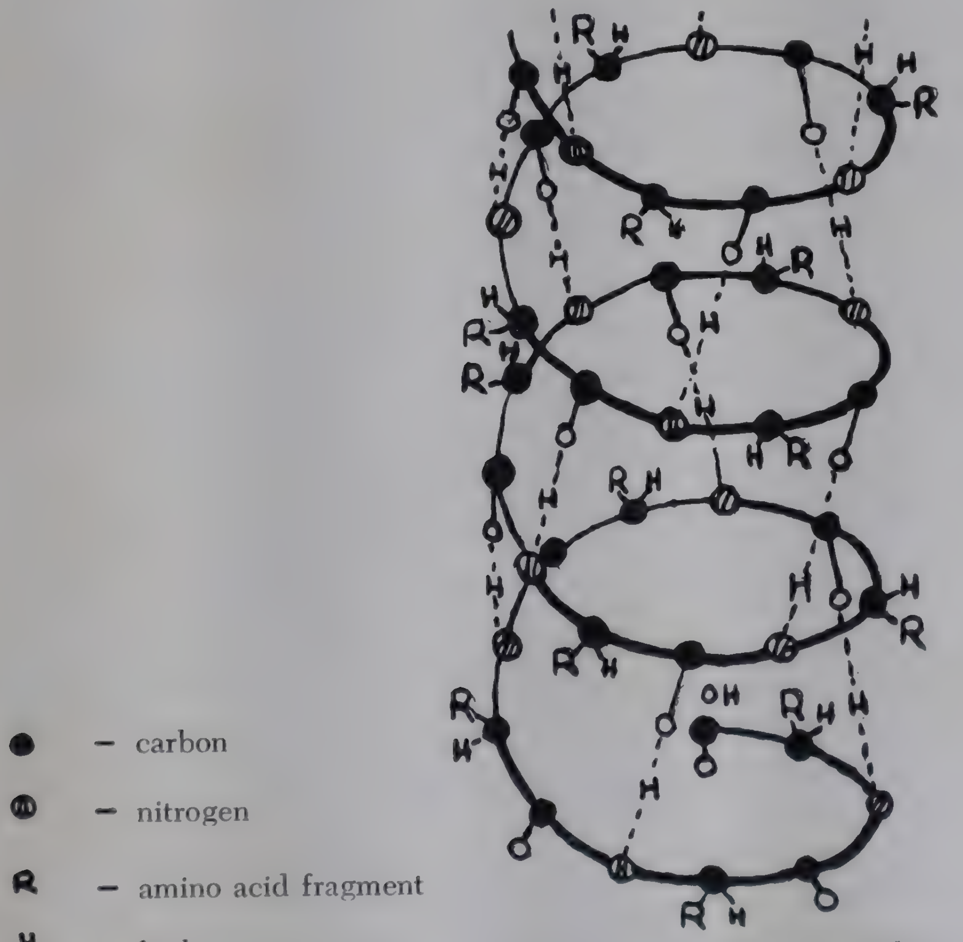
Fig. 7. Geometry of Molecules

receiving a share of two more electrons from two hydrogen atoms. One possible configuration of the eight outermost electrons is to have one pair of electrons distributed about the nucleus in spherically symmetrical fashion and the other three pairs directed outwards from the nucleus at right angles to one another. Another possible configuration, which requires more energy is one in which the four outermost electron pairs are directed towards the corners of a regular tetrahedron, forming an angle of approximately 109 degrees between electron pairs. The actual angle between hydrogen atoms is then a compromise between the 90 degree angle dictated by lowest electron energy and the repulsion energy between the hydrogen nuclei, which decreases as the angle between hydrogens increases. In a similar fashion, the already high symmetry of the ammonia molecule is made even more so when one considers that in this molecule there exists a pair of electrons belonging to the nitrogen atom which is directed up from the pyramid apex towards one corner of the tetrahedron, of which the other three are occupied by hydrogen atoms. The experimentally determined angle between hydrogen atoms is 109 degrees, which is within one degree of the angle expected from a tetrahedral configuration.

The quantum mechanical theory of electron orbitals in atoms and molecules has given an essentially complete theoretical basis for the spatial orientations of electron distribution, and hence of bond angles and molecular geometry. In the case of molecules whose atoms are connected by single bonds, the bond angles, including those of unshared electron pairs, will lie between 90 degrees and 109 degrees. If double bonds are involved, the bond angles involving those atoms will generally be in the neighborhood of 120 degrees.

For organic molecules, the most important bond angle is the tetrahedral angle of 109 degrees. In the case of small organic molecules, containing only a few carbon atoms, the number of molecular shapes or configurations is very small. On the other hand, in the case of large molecules, involving hundreds or even thousands of carbon atoms, the number of possible molecular configurations becomes large enough and varied enough to pose problems of considerable chemical and biological interest.

Consider now, a large molecule in which adjacent atoms in a chain are connected at tetrahedral angles (109 degrees). These molecules may be either essentially straight chains, erratic chains, or coiled up chains. In the case of large molecules, these possible differences in configuration can result in profound changes in chemical and biological behavior. A protein molecule which consists of thousands of small molecular building blocks, called amino acids, is evidently a type of molecule which is capable of assuming a large variety of configurations, such as described above. A currently accepted structure for certain protein molecules is a



Courtesy of Thorpe

Fig. 8. Helical Configuration of Protein Molecule

spiral structure (see Figure 8). This however, is only one of the many possible configurations which this large molecule can assume. The process of protein denaturation, which is a profound change in properties which can be brought about by heat, the addition of alcohol and other chemicals, consists of a change in the configuration of this molecule and not at all of a change in the conventional chemical sense. A number of other processes of biological importance involve changes in the configuration of large molecules.

Molecular Motions

Molecules execute a variety of motions, including translation, rotation, and vibration. In translation the molecule as a whole is moving in some particular direction. In the gaseous state this motion is comparatively unimpeded; a molecule may move for a distance of many thousand molecular diameters before its direction of motion is changed by collision with another gas molecule. When such a molecule collides with the wall of the container, it exerts a force on the wall. The sum of these many microscopic impacts is the pressure which the gas exerts.

In liquids, the distance between molecules is reduced to a value roughly equal to the molecular diameter and a molecule cannot travel unimpeded for any distance at all. The molecules do, however, manage to change their location in the following manner. The arrangement of molecules in a crystal consists of a nearly regular arrangement of molecules in space, with only an occasional imperfection, such as an unoccupied lattice position or an extra molecule crowded in. In liquids, the density (molecules per unit volume) is almost always slightly less, and this decreased density is accompanied by the presence of a greater number of unoccupied lattice positions. When the number of unoccupied lattice positions exceeds a critical value, the overall orderly arrangement of molecules breaks down and the material loses its rigidity; it becomes liquid. An observer sitting on any particular molecule would be surrounded by neighboring molecules arranged in the nearly regular crystalline array. But those molecules located further out do not quite fit into the pattern any more, because of the presence of vacant positions. A molecule may now move into a vacant position by squeezing through the immediate neighbor shell. This process of squeezing through requires extra energy to overcome the repulsion of the neighbors, and thus will only occur if the given molecule acquires enough energy momentarily. The energy is acquired, and lost again, by the recurring collisions between the molecules. The dynamic state of a given molecule in a liquid may thus be described as being confined in a particular location for a time by the repulsive forces of neighboring molecules which form a “cage.” The molecule bounces back and forth in this cage, continually exchanging energy with its neighbors and occasionally, when it happens to have the required excess energy slipping out of the cage, or squeezing between the neighbors, to a vacant place outside the cage. Compared to the gaseous state, then, the translational (or diffusion) process is considerably slower.

The state of affairs in solids is much like that in liquids. Because of the presence of vacancies in the lattice there are alternate locations available into which atoms or molecules can move. Thus even a solid-appearing material such as a metal can be permeated by other atoms and molecules. For example a platinum container will not retain hydrogen gas, it diffuses right through the walls, and the same is true of attempting to store sodium in some metallic containers at high temperatures. Also, radioactive tracer experiments have revealed a continuing interchange of lattice positions among the atoms of many solid materials.

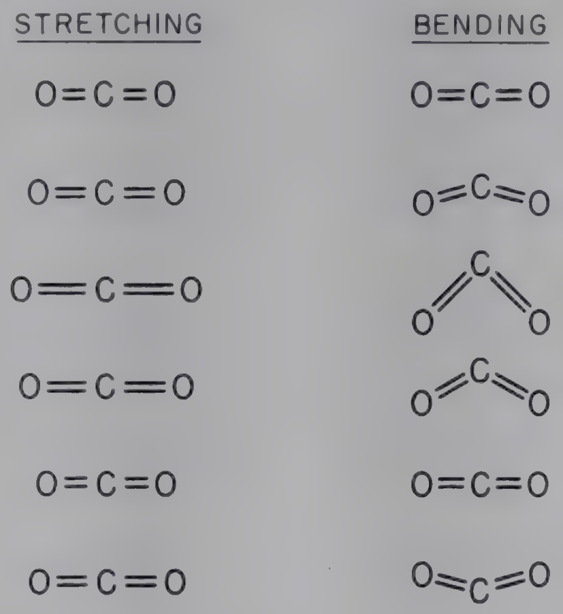


Fig. 9. Molecular Stretching and Bending Vibrations Schematically Shown in Series of "Movie Stills"

Molecular rotations again are comparatively unimpeded in the gaseous state and are more or less restricted in the liquid and solid states. The rotations are also motions which are quantized. The wave length of light absorbed and emitted in transitions among the various rotational states falls into the radiofrequency and the far infrared heat wave regions.

Molecular vibrations are, like translations, of considerable importance in processes of radiation damage and chemical change. In the gaseous state, for example, the collisions among the constantly moving molecules provide a means for transferring energy between molecules. The vibrations of the molecules, which may be pictured as repeating distortions of the atomic arrangement of a molecule are of importance in processes in which a molecule breaks apart into fragments or, conversely, two or more fragments collide and establish a new molecule.

The molecular vibrations are conveniently classed as "stretching" and "bending" vibrations, the nature of which is illustrated in Fig. 9. The significance for chemical change is illustrated by the vibrations of the ethane molecule, one vibrational motion of which is a repeated stretching and contraction of the carbon-carbon bond. As this vibration becomes more violent, this bond stretches and when a sufficient amount of vibrational energy is concentrated in this motion the bond will rupture.

Free Radicals

A class of molecules of considerable importance for radiation phenomena are the free radicals. A free radical is an extremely reactive molecular species which has a tendency to dimerize. The simplest free radical is the hydrogen atom, which has very high tendency to react with other molecules, and dimerizes to form a hydrogen molecule, H_2. Other examples of interest include the radical $\cdot OH$, which can dimerize to form hydrogen peroxide, H_2O_2, or combine with a hydrogen atom to form water, methyl radicals $\cdot CH_3$, amine radicals $\cdot NH_2$, and a variety of atoms such as oxygen and iodine.

A figurative process for generating free radicals is to take a stable molecule and remove an atom or group of atoms from it, thereby breaking a bond. In this way a cause for the reactivity of this type of species becomes apparent. Generally speaking, free radicals always possess an unpaired or "odd" electron which is capable of forming another chemical bond. It may be seen from the preceding discussions on atomic structure and on valence that the number of electrons possessed by stable molecules is even, with only a very few exceptions, out of thousands of cases.

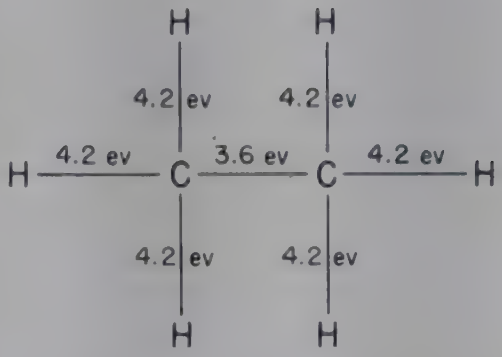
Free radicals may be generated in practice by any means which will provide enough vibrational energy to cause a bond rupture. Processes of this type include heating, absorption of light of the proper frequency, and excitation of molecules by collision with any types of particles—nuclear, atomic, electronic, ionic, or molecular—which can supply a sufficient amount of energy.

Bond Energies

The amount of energy that must be supplied to a given chemical bond in order to break it is termed the bond energy. This quantity is of importance in connection with the detailed manner in which molecules will decompose after they have been energized by any of the above approaches. In molecules containing large numbers of atoms, the possible ways of decomposing increase rapidly with molecular size and a knowledge of bond energies is exceedingly useful in understanding and, some-

Fig. 10. Comparative Strength of Bonds in Ethane

Note: The different types of bonds in a given molecule are generally not of equal strength.



times, predicting the detailed behavior of molecules under excitation. The ability to sketch out, at least approximately, the most likely decomposition processes gives information on the nature of active species produced by radiation and thus provides some insight into the complicated chain of events leading to the overall gross effects of radiation. The energies of various bonds in a given molecule are not all equal. Further, the energy of a bond connecting a given pair of atoms such as C–C, or C–H, or O–H depends in first approximation on the nature of the two atoms directly concerned but will vary slightly, depending on what atomic groupings are otherwise attached to one or both members of the pair. It is seen from the accompanying Table 7 that most bond energies range from perhaps

Table 7

DISSOCIATION ENERGIES OF MOLECULAR AND IONIC BONDS

	Molecules[1]		Ions[2, 4]	
Chemical Bond[3]	*D*, kcal	*D* Electron Volts, ev	*D*, kcal	*D*, ev
H—H	103.24	4.5	61	2.6
CH_3—H	101	4.4	31	1.3
C_2H_5—H	96	4.2	25	1.1
C_6H_5—H	102	4.4	110	4.8
NH_2—H	102	4.4	119	5.2
HO—H	116	5.0	146	6.3
HS—H	~90	3.9	109	4.7
H_3C—CH_3	83	3.6	53	2.3
$H_2C{=}CH_2$	~125	5.4	203	8.8
$HC{\equiv}CH$	~166	7.2	251	10.9
CH_3—CHO	~75	3.3	—	—
HOC—H	~78	3.4	34	1.5
C_2H_5—OH	90	3.9	—	4.0
C_2H_5O—H	19	0.8	91	—
$RC{=}O$	~160	6.9	—	—
RC—NR_2	~56	2.4	—	—
F_3C—F	~94	4.1	—	—
Cl_3C—Cl	70	3.0	—	—
Br_3C—Br	~50	2.2	—	—

---

[1] Cottrell, T. L. 1954. The Strengths of Chemical Bonds. Academic Press, Inc., New York.

[2] Field, F. H., and Franklin, J. L. 1957. Electron Impact Phenomena and the Properties of Gaseous Ions. Academic Press, Inc., New York.

[3] Particular bond in question indicated by dash.

[4] Refers to same chemical species bearing one positive charge.

one to five electron volts, and from Fig. 10 that the various bonds in a given molecule may differ in energy by only a few tenths of an electron volt. These small differences, however, exert a major effect on molecular decomposition paths. The concept of bond energy applies equally well to ionized molecules. A number of values are given in the same table. Generally, the bond energies of ions are substantially less than those of the corresponding neutral atom because the electron removed in the ionization process contributed to the bonding in the neutral molecule.

BIBLIOGRAPHY

Bohr, N. 1934. Atomic Theory and the Description of Nature. Cambridge University Press, Cambridge, England.

Bohr, N. 1958. Atomic Physics and Human Knowledge. John Wiley and Sons, New York.

Cottrell, T. L. 1958. The Strength of Chemical Bonds. Second Ed., Academic Press, New York.

Coulson, C. A. 1952. Valence. Oxford University Press, London, England.

Eyring, H., Walter, J., and Kimball, G. F. 1944. Quantum Chemistry. John Wiley and Sons, Inc., New York.

Fano, U., and Fano, E. 1959. Basic Physics of Atoms and Molecules. John Wiley and Sons, Inc., New York.

Pauling, L. 1940. The Nature of the Chemical Bond. Cornell University Press, Ithaca, New York.

Pritchard, H. O. 1953. The determination of electron affinities. Chem. Revs. 52, 529.

Rice, F. O., and Teller, E. 1949. The Structure of Matter. John Wiley and Sons, Inc., New York.

Syrkin, Y. K., and Dyatkina, M. E. 1950. Structure of Molecules. Interscience Publishers, Inc., New York.

Wells, A. F. 1950. Structural Inorganic Chemistry. 2nd edition. Oxford University Press, London, England.

CHAPTER 4

Chemical Reactions Induced by Radiation

The Nature of Chemical Reactions

To undergo chemical reaction, molecules must come into close proximity and form the product molecules by breaking some chemical bonds and establishing certain new ones. At the same time the arrangement of atoms continuously changes from that of the original molecules to that characteristic of the product molecules. This rearrangement of chemical bonds and atomic configuration generally requires more energy than the molecules normally possess, and in this transformation there is a stage at which it can proceed further without any more energy being supplied. This configuration of maximum energy is called the activated complex. The rate at which the chemical change can occur is proportional to the concentration of activated complexes, which depends on the excess energy they must have and on the likelihood of forming the activated configuration. The greater the excess energy required, the less the concentration of the activated complex. As the temperature of the system is raised, the average energy of the molecules is raised and the concentration of activated complexes increases. This is in accord with experience according to which chemical reactions occur more rapidly at higher temperatures.

For a set of molecules which can undergo a variety of possible reactions, there is a different activated complex for each reaction, with its own characteristic structure and energy. In a container not subject to irradiation the relative probability of occurence of these various activated complexes is governed by statistical laws in which the external factors of temperature and pressure offer the only means of direct control over the outcome of the reactions. The principal chemical reactions that do in fact occur are those whose activated complexes are formed in the highest concentration. This will be the case primarily for those activated complexes that have the smallest excess energy requirement. Frequently, only one activated complex is present in significant concentration and the resulting chemical reaction is specific.

However, if temperature and pressure were the only means of control, our chemical technology would not be as fully developed as it is today. Chemical technology was greatly expanded by the discovery of catalysts. The action of catalysts is to allow the easier attainment of totally different activated complexes which yield a given end product. Although each reaction involves a different corresponding activated complex, the converse is not true. A number of activated complexes may produce the same end product. The introduction of catalysts and catalytic surfaces permits an entirely new class of activated complexes in which the atoms and molecules of the added catalyst play a role.

Initiation of Chemical Reaction with Radiation

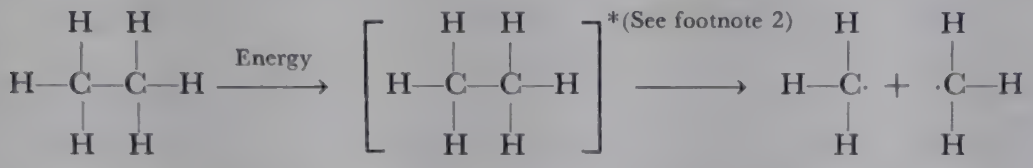
Radiation produces a variety of chemical species such as ions, radicals, and excited molecules. These species undergo further reactions among themselves and with their environment to form radiation products. Among the fragments that may be found are many that require such great amounts of energy in order to be formed that their formation in useful amounts without radiation may be difficult or even impossible by conventional techniques. However, once obtained, the species may now form activated complexes which will lead to products in the same way and following the same laws as is the case in conventional chemical change. The new feature of radiation chemistry is the greater flexibility of obtaining reactive species in a variety of surroundings and over wider ranges of temperature and pressure. Further, the very fact that irradiation of a molecule can produce pronounced changes in a single step offers promising possibilities of eliminating numerous steps in chemical processing which otherwise may be required to effect a particular chemical change. Another possibility is that of creating chemical changes which are at present very difficult to carry out. It should be remembered that the radiation part of radiation chemistry is the new way of creating reactive species. The chemistry part of radiation chemistry, which consists of the rules of formation and of reaction of these species, is the same set of rules that govern conventional chemical changes. Some useful chemical reactions induced by radiation are given below:

Formation of lindane, an insecticide, from benzene and chlorine.
Direct production of phenol (carbolic acid) from benzene, water, and oxygen.
Direct oxidation of hydrocarbons leading to the formation of aldehydes, ketones and acids.
Production of ethylene glycol (an anti-freeze) from methanol (wood alcohol).
Formation of aniline, a dye-intermediate, from benzene and ammonia.
Fixation of nitrogen directly.
Formation of alkyl sulfonates (detergents) from hydrocarbons and sulfur dioxide.
Formation of ascorbic acid (vitamin C).
Irradiation of yeast to enhance the production of ergosterol, a precursor of vitamin D.

Decomposition of Excited Molecules and Ions

A type of chemical reaction which is of importance in radiation effects is the decomposition of excited molecules. When a molecule or ion has been given sufficient excitation energy by collision or irradiation to rupture one or more chemical bonds, this process can take place irrespective of the proximity of other molecules. However, there is still a well defined activated complex, in which the distribution of the energy among the internal degrees of freedom of the molecule plays a central role.

For example in the decomposition of ethane into two methyl radicals[1]



it is necessary that the molecule possess at least 3.6 electron volts of excitation energy, which is the energy of the carbon-carbon bond. Further, this minimum amount must be concentrated in this particular bond in order that it vibrate with sufficient amplitude to rupture this bond.

If more than four electron volts are available, the molecule has enough energy to rupture either the carbon-carbon bond or one of the C–H bonds (energy = 4.2 ev).

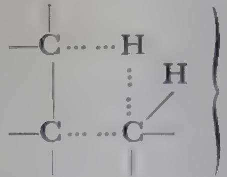
The extent to which either of these reactions occurs is determined primarily by the activation energy. The excitation energy in the molecule is constantly being redistributed among the degrees of freedom in an essentially random fashion. At some time instant a sufficient amount of energy will be concentrated in the proper degree of freedom (or chemical bond) and the molecule can decompose. The probability of this depends sensitively on the fraction of the available energy which must be concentrated in the bond. Thus as the amount of available energy is increased,

---

[1] The symbols below are used to identify ions, radicals, activated complexes, and excited species.

H^+ ion

$\left.\begin{array}{l}\cdot OH\\ \cdot CH_3\\ \cdot C_2H_5\end{array}\right\}$ free radicals

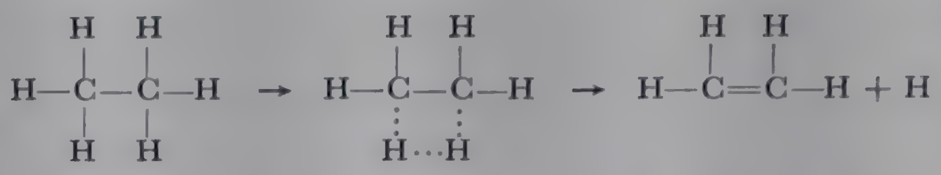


Activated Complex (dotted line indicates transitory bonds as occur in activated complex)

[2] $[C_2H_6]^*$ excited chemical species.

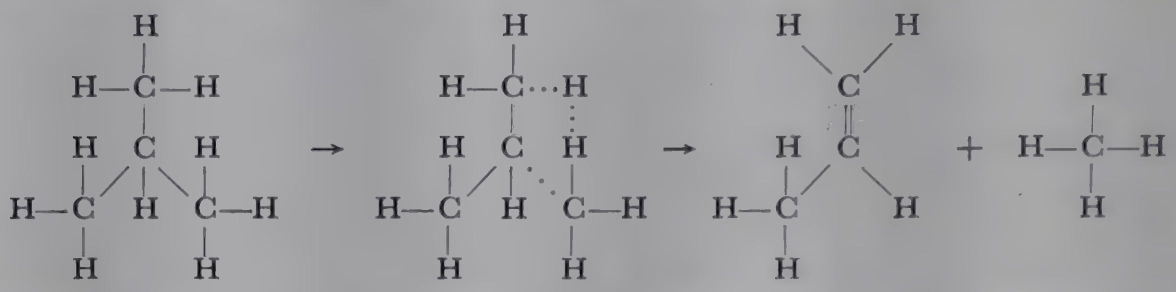
the rate of decomposition increases and, at any energy, the reaction with the lower activation energy will be favored.

The decomposition reactions of the excited molecules are by no means confined to simple bond rupture processes such as those above. There are many cases in which bonds rearrange in the course of decomposition, leading to formation of smaller molecules rather than radicals. Typical examples are the dehydrogenation of ethane to pure ethylene and hydrogen molecules



$$\mathrm{H{-}\overset{H}{\underset{H}{C}}{-}\overset{H}{\underset{H}{C}}{-}H} \rightarrow \mathrm{H{-}\overset{H}{\underset{\underset{H}{\vdots}}{C}}{-}\overset{H}{\underset{\underset{H}{\vdots}}{C}}{-}H} \rightarrow \mathrm{H{-}\overset{H}{C}{=}\overset{H}{C}{-}H + H}$$

and the decomposition of isobutane to form methane and propylene.



Other reactions include the removal of water, ammonia, hydrogen chloride, ethane, and hydrogen sulfide from a variety of molecules and ions. It is to be re-emphasized that these reactions occur each in one single step.

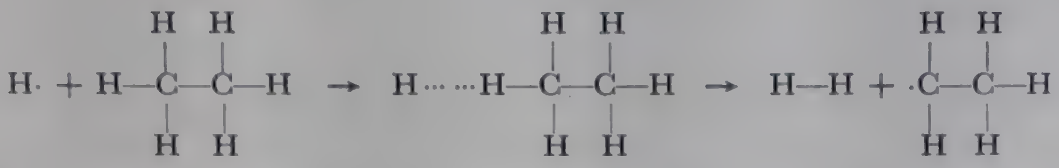
The bond rupture reactions differ from the reactions involving loss of molecules in one important respect. In the course of bond rupture, the molecule loosens whereas in the latter reactions additional transitory bonds are established which form additional restrictions on the internal rotations and vibration of the molecules. These effects lead to some changes in the probability of forming an activated complex, as determined by energy alone. The bond rupture processes are somewhat more favored than the loss of neutral molecules.

Reactions of Radicals

Free radicals such as are formed by the decomposition of excited molecules undergo a variety of reactions of four types which are of interest here:

a. abstraction
b. recombination
c. disproportionation
d. polymerization

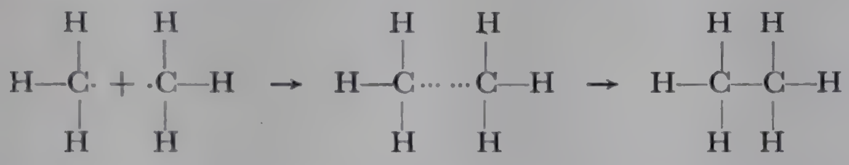
In an abstraction reaction a free radical collides with another molecule and removes an atom. As a result of this, the radical generally becomes a normal molecule and the other molecule a free radical. A simple example of this is hydrogen abstraction. A hydrogen atom, possibly formed by decomposition of excited ethane, may abstract a hydrogen atom from another ethane molecule, forming molecular hydrogen and an ethyl radical.



$$\mathrm{H\cdot + H{-}CH_2{-}CH_2{-}H \rightarrow H\cdots H{-}CH_2{-}CH_2{-}H \rightarrow H{-}H + \cdot CH_2{-}CH_2{-}H}$$

The irradiation of many organic systems leads to the production of hydrogen gas. This is formed both by loss of molecular hydrogen and formation of hydrogen atoms which subsequently undergo the above type of abstraction reaction. The course of abstraction reactions is governed energetically by the strength or energy of the bond which is being formed as compared to that of the bond being broken.

The recombination of free radicals is a process converse to the decomposition of excited molecules.



$$\mathrm{H{-}CH_2\cdot + \cdot CH_2{-}H \rightarrow H{-}CH_2\cdots CH_2{-}H \rightarrow H{-}CH_2{-}CH_2{-}H}$$

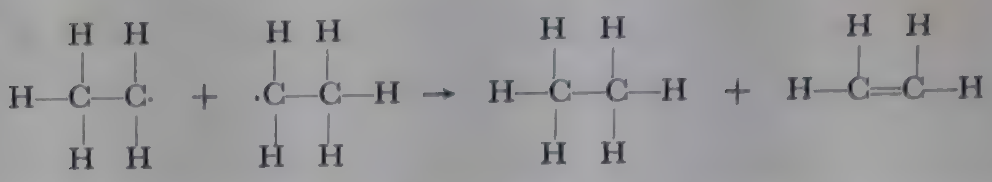
The activated complex formed by two recombining methyl radicals is identical with that for the reverse process, decomposition of excited ethane to form two methyl radicals. While in the decomposition reaction it is necessary to concentrate the energy in the carbon-carbon bond, in the recombination reaction this energy is concentrated there in the course of the process and it is necessary to redistribute or remove some of this energy in order that the reaction can proceed to completion. In the case of small molecules this requires that a third molecule collide with the activated complex to remove some of the energy. Large molecules may effectively recombine by internal redistribution of the energy.

Another recombination reaction of importance in radiation chemistry is the formation of hydrogen peroxide from hydroxyl radicals.

$$\mathrm{HO\cdot + \cdot OH \rightarrow H{-}O\cdots O{-}H \rightarrow H{-}O{-}O{-}H}$$

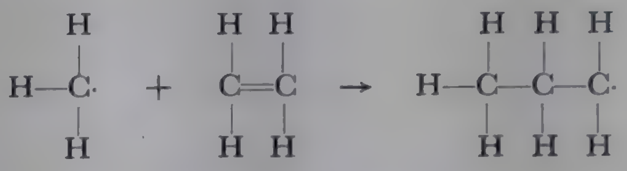
In this reaction the recombination energy is dissipated by collision with surrounding water molecules.

In a disproportionation reaction two radicals form normal molecules by transfer of an atom or group of atoms.



$$\mathrm{H_3C{-}CH_2\cdot + \cdot CH_2{-}CH_3 \rightarrow H_3C{-}CH_3 + H_2C{=}CH_2}$$

A polymerization reaction occurs if a free radical can combine with another molecule without losing its free radical characteristics. For example a methyl radical can combine with an ethylene molecule to form a propyl radical.



$$\mathrm{H_3C\cdot + H_2C{=}CH_2 \rightarrow H_3C{-}CH_2{-}CH_2\cdot}$$

This radical can in turn combine step by step with other ethylene molecules to form still larger molecules.

$$\begin{array}{rcl}
\text{radical} + \text{monomer} & \rightarrow & \text{dimer radical} \\
\text{dimer radical} + \text{monomer} & \rightarrow & \text{trimer radical} \\
\vdots \qquad\quad \vdots & \vdots & \vdots \\
n\text{-mer radical} + \text{monomer} & \rightarrow & (n+1)\text{-mer radical}
\end{array}$$

This process can continue until the radical reactivity is lost by a recombination or disproportionation reaction with another radical.

Reactions of Ions

Atomic and molecular ions are reactive species which are produced by radiation along with radicals and excited molecules. The types of reaction which these species can undergo are to some extent similar to those discussed in the preceding sections but present enough new features to warrant separate discussions (Rosenstock 1952, 1959). The following four types of reactions are of importance in discussing radiation effects:

a. Decomposition of excited ions
b. Charge transfer
c. Ion-molecule reactions
d. Ion-electron recombination

A large excited polyatomic ion such as $C_2H_5OH^+$ will decompose to form fragments in the same manner as an excited molecule. The products of the decomposition will always include one charged fragment. The other fragment may be either a radical or a normal molecule. When such an ion decomposes the charged fragment is the one which has the lower ionization potential. Thus, for example, an ethane ion may decompose in the following way:

$$[C_2H_6^+]^* \rightarrow C_2H_4^+ + H_2$$

but not

$$[C_2H_6^+]^* \rightarrow C_2H_4 + H_2^+$$

Another example is:

$$[C_2H_5OH^+]^* \rightarrow C_2H_5^+ + \cdot OH$$

but not

$$[C_2H_5OH^+]^* \rightarrow \cdot C_2H_5 + OH^+$$

The relevant ionization potentials are given in Table 8.

TABLE 8

SOME IONIZATION POTENTIALS[1]

Chemical Species	ev
C_2H_4	10.5
H_2	15.4
$\cdot C_2H_5$	8.7
$\cdot OH$	13.7

[1] Franklin, J. L., and Field, F. H. (1957).

The decomposition reactions of these ions include both bond rupture and loss of normal molecules. As before, the loosening or restriction of the internal motions of the molecular ion in the activated complex modifies the course of the decomposition reactions.

In a charge transfer reaction a positive ion approaches a neutral molecule and removes an electron from it. In order for this reaction to occur it is necessary that the ionization potential of the molecule be equal to or less than the energy liberated by the capture of the electron by the ion. This latter quantity is the ionization potential of the neutral counterpart of the approaching ion. If the ionization potential of the target molecule is less than the energy liberated, the excess will go into internal excitation of the target, resulting in decomposition. The following are examples of these reactions.

$$A^+ + CH_4 \rightarrow A + CH_3^+ + H\cdot$$
$$Kr^+ + CH_4 \rightarrow Kr + CH_4^+$$

and the relevant energies are given in Table 9.

TABLE 9

ENERGY REQUIREMENTS FOR SOME IONIZATION AND DISSOCIATION PROCESSES

Process	Energy Required, ev
$A \rightarrow A^+ + e$	15.7
$Kr \rightarrow Kr^+ + e$	14.0
$CH_4 \rightarrow CH_4^+ + e$	13.1
$CH_4 \rightarrow CH_3^+ + \cdot H + e$	14.4

It is seen that capture of an electron by a krypton ion, Kr^+, liberates only enough energy to ionize methane, with an argon ion, A^+, however, there is enough energy available to dissociate the methane into a methyl ion and a hydrogen atom. These charge transfer reactions can occur at separations of several molecular diameters, greater than the maximum separations at which the usual chemical reactions occur.

Ion-molecule reactions include a wide variety of processes including disproportionation, abstraction, and polymerization reactions. Some recently observed reactions include:

$$H_2^+ + O_2 \rightarrow HO_2^+ + H\cdot$$
$$CH_4^+ + CH_4 \rightarrow CH_5^+ + \cdot CH_3$$
$$C_2H_2^+ + C_2H_4 \rightarrow C_3H_3^+ + \cdot CH_3$$

These reactions are important intermediate steps in many radiation-induced chemical reactions. They differ from neutral molecule and radical reactions in two important respects. In comparison to the reactions of neutral species,[1] the rates of these processes are much more rapid. The reason is that the frequency of collisions between ions and molecules is much larger because of the much greater attractive forces between the reactant species. Thus if a molecule passes an ion with a minimum separation of several molecular diameters, the attractive forces will draw the two reactants together to the point where they can form an activated complex. The forces between neutral species are of much shorter range. A second important difference exhibited by these processes is that they can only be intermediates in an overall chemical change since eventually the positive ion is neutralized by an electron.

The details of ion-electron recombination reactions are only very incompletely understood. On recombination the neutral fragment is highly excited. This excitation may be carried away by collision with another molecule or it may lead to decomposition of the neutralized ion. A third possibility is that in the recombination process the energy is liberated as light. Up to the present no detailed studies are available on the relative importance of these processes in the case of molecules. It is possible that decomposition is the major effect of ion-electron recombination in most molecules.

BIBLIOGRAPHY

DAINTON, F. S. 1956. Chain Reactions: An Introduction. John Wiley and Sons, Inc., New York.

---

[1] The word species here and in the text to follow is used in the broad chemical sense of any combination of atoms and electrons in any manner or form. The term includes atoms, molecules, radicals, ions, excited atoms, and excited molecules either singly or in large quantity.

Franck, J., and Platzman, R. L. 1954. Physical principles underlying photochemical, radiation–chemical and radiobiological reactions. Radiation Biology. Editor, A. Hollaender. Vol. I. Chapter 3. McGraw-Hill Book Company, Inc., New York.

Franklin, J. L., and Field, F. H. 1957. Electron Impact Phenomena and the Properties of Gaseous Ions. Academic Press, New York.

Glasstone, S., Laidler, K., and Eyring, H. 1941. The Theory of Rate Processes. McGraw-Hill Book Co., Inc., New York.

Laidler, K. J. 1955. The Chemical Kinetics of Excited States. Oxford University Press, London, England.

Massey, H. S. W., and Burhop, E. H. S. 1952. Electronic and Ionic Impact Phenomena. Oxford University Press, London, England.

Rosenstock, H. M., Wallenstein, M. B., Wahrhaftig, A. L., and Eyring, H. 1952. Absolute rate theory for isolated systems and the mass spectra of polyatomic molecules. Proc. Nat. Acad. Sci. 38, 667–678.

Rosenstock, H. M., et al. 1959. Ion molecule reactions. U.S.A.E.C. Contract Rept. No. JL1-650-3-7.

Semenov, N. 1958. Some Problems of Chemical Kinetics and Reactivity. Pergamon Press, New York.

Steacie. 1954. Atomic and Free Radical Reactions. Second Ed. Reinhold Publishing Corp., New York.

Trotman-Dickenson, A. F. 1955. Gas Kinetics. Butterworths Scientific Publications, London, England.

Wallenstein, M. B., Rosenstock, H. M., and Johnston, W. H. 1959. Report on Fundamental Considerations of Radiation Technology, Parts I and II. JLI-650-3-3, JLI-650-3-5, AEC Contract 4T(11-1)-650.

Walling, C. 1957. Free Radicals in Solution. John Wiley and Sons, Inc., New York.

CHAPTER 5

Physical Effects of Radiation (or Interaction of Radiation with Matter)

When radiation impinges on a target material, it brings about physical, chemical, and biological effects by transferring its energy to the material. The energized regions of the material, which are microscopic in size, then undergo a variety of transformations which ultimately lead to permanent changes in the physical, chemical and biological characteristics of the material. The complex sequence of events leading to these changes can be divided into three steps (Magee 1953):

a. the transfer of energy from radiation to the target material
b. the transformation of the energized regions into particular excited and active atomic and molecular species which produce the changed characteristics of the material
c. the reaction of these species among themselves and with their environment which lead to these changes.

For any particular energized region, these three steps are consecutive and have a duration of roughly 10^{-17}, 10^{-13} to 10^{-10}, and 10^{-10} seconds and longer (up to seconds or minutes), respectively. In this chapter we shall be concerned with the first two steps.

The interaction with matter will be discussed separately for four classes of radiation:

a. high energy electrons
b. atomic radiation, i.e., protons, alpha particles of fission fragments
c. electromagnetic radiation, i.e., X-rays and gamma rays
d. neutrons

Interaction of High Energy Electrons with Matter

When a high energy electron travels through matter it can lose energy by collision with electrons of the target material and by emitting radiation called "bremsstrahlung." In the collision process the electrons of the material are excited or completely ejected from the atom at the expense of some of the energy of the incident high energy electron. If in this process an electron is ejected, there is no way of distinguishing the ejected electron; the faster of the two electrons may be the incident high energy electron which has lost a small amount of energy or, just as well, the target electron which has received most of the energy in the collision. The slower one of the electron pair resulting from a collision is arbitrarily called the ejected electron. The amount of energy imparted to the ejected electron may range from zero up to one half of the energy of the incident electron. However, ejection with high energy is less probable than ejection with low energy (see Fig. 11).

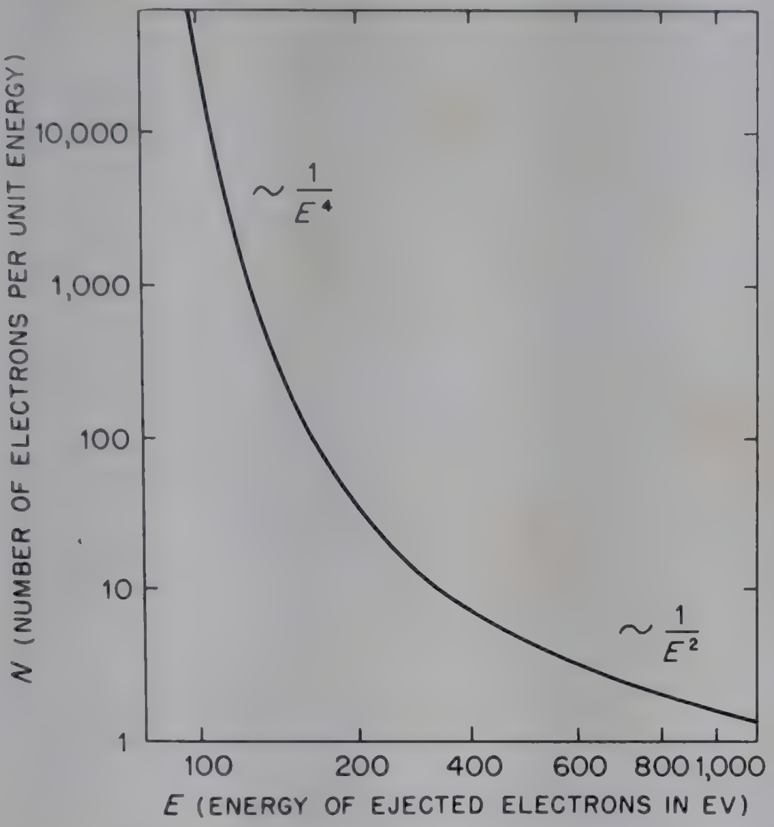


FIG. 11. DISTRIBUTION OF ENERGY OF ELECTRONS EJECTED BY COLLISION PROCESS

The entire sequence of collisions leading to dissipation of the incident energy may be described as a cascade process. A single high energy electron loses its energy by exciting some electrons and by liberating a number of secondary electrons. These secondary electrons have a variety of energies, in some cases high enough to eject a considerable number of additional electrons, and some of these third generation electrons can in turn eject further electrons. Theoretical calculations and measurements indicate that on the average about thirty electron volts of energy are dissipated per electron ejected by the incident radiation. Comparing this to the energy required to ionize atoms and molecules, about 10 to 15 volts, it follows that roughly half of the incident radiation energy is expended in ionization and half in excitation of the target material.

For very high energy electrons, above about ten million electron volts, the dominant mechanism of energy loss is bremsstrahlung (see Fig. 12).

In this process an electron is deflected in its path by the electric field of a nucleus of the target material. This deflecting force of the nucleus produces an acceleration of the electron, in consequence of which the electron emits electromagnetic radiation and loses energy. This radiation may have any energy from zero up to the energy of the electron less 0.51 Mev, and is fairly uniformly distributed over the entire range. Thus a 10 Mev electron may emit bremsstrahlung having an energy up to 9.49 Mev which corresponds to a high energy gamma ray. Part of the energy dissipation problem for high energy electrons thus involves an understanding of the interaction of high energy electromagnetic radiation with matter. This will be discussed shortly.

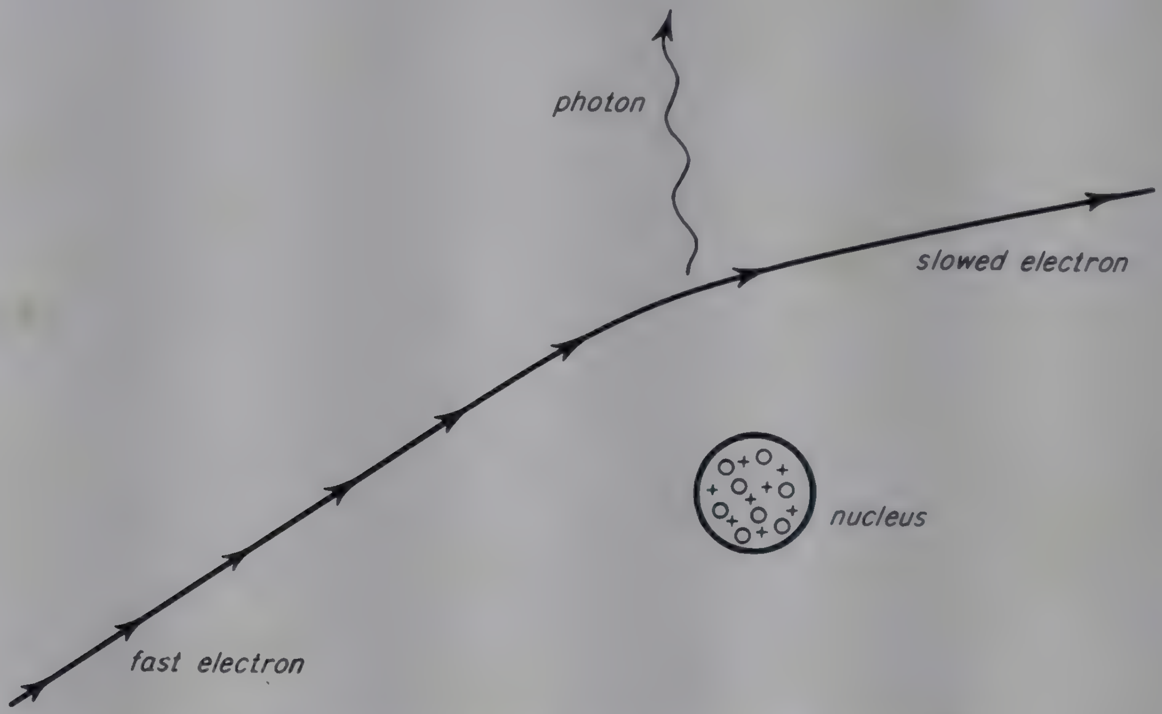


Fig. 12. Bremsstrahlung or Deceleration Radiation

Besides undergoing energy-dissipating or inelastic collisions with the target electrons and nuclei a high energy electron may also undergo elastic collisions with the target electrons and nuclei. In these collisions the electron does not lose energy but is deflected in its path, occasionally to large angles. Because of this scattering the path of a high energy electron in a target material is not at all a simple straight line but an irregular zig-zag path. Further, this irregularity creates some difficulty in describing the depth to which high energy electrons can penetrate into material. For example, an electron may require ten centimeters of path length to dissipate its energy in a given target material. However, the irregular nature of the path leads to a considerable (and variable) difference between this path length and the depth of penetration.

Interaction of Atomic Radiation with Matter

The mechanisms of energy loss of atomic particles—protons, alpha particles and fission fragments—are somewhat similar to those of the electrons. The principal mechanism is inelastic collisions with the electrons of the target material, leading to ionization and excitation. The partition of energy between excitation and ionization is roughly equal, as was the case with electrons, and, again, the energy per ionization is about thirty electron volts. The rate of energy loss for these atomic particles, however, is considerably greater than for electrons, increasing in the order protons, alpha particles and fission fragments. The reason for this is that an atomic particle of a given energy travels much more slowly than an electron of the same energy, and the probability of an energy dissipating collision increases as the velocity decreases. The large mass difference between these particles and the electrons leads to other differences in behavior. Because of the heavier mass, an encounter with the field of a nucleus leads to much smaller acceleration, so that bremsstrahlung energy loss is negligible. Further, in an elastic collision with the electrons of the target material, the mass of the atomic particles is so much greater that they experience almost no deflection. Thus the paths of these particles are very nearly straight lines.

Effects of Gamma Rays

Gamma rays energize target materials primarily by three types of mechanisms: the photoelectric effect, the Compton effect, and electron-positron pair production. The three mechanisms are of different relative importance, depending on the atomic number of the target material and the energy of the gamma photon. For example, in a low atomic number material such as aluminum ($Z = 12$) the photoelectric effect is important at energies below about 0.05 Mev, above this energy the Compton effect predominates and, above 15 Mev the principal effect is pair production. In a heavy element such as lead ($Z = 82$), the respective limits are below 0.5 Mev for the photoelectric effect, above this the Compton effect and, above 5 Mev, pair production.

In the photoelectric effect a single gamma photon is completely absorbed by an atom with the ejection of an electron. The electron is ejected with a kinetic energy equal to the energy of the photon less that required to overcome the binding energy of the electron. It is characteristic of this process that preferably the most tightly bound electrons of the atom are ejected if the photon energy is sufficient. If not, then the next most tightly bound ones, and so on. Thus this process results in the formation of ionized species which are highly excited electronically.

The Compton effect is a manifestation of the particulate nature of electromagnetic radiation. Just as particles have wave properties, so do electromagnetic waves have particle properties. A photon has a momentum and can undergo collisions with particles. In this case the particles are electrons. The result of such a collision is the transfer of energy and momentum to the electron at the expense of the energy and momentum of the photon. The energy transferred to the electron may be an appreciable fraction of the photon energy. For example, a 1.3 Mev gamma photon such as is emitted by cobalt-60 may produce Compton electrons having energies up to 1.08 Mev. In this collision process the photon does not vanish, but is degraded to a photon of lower energy which may interact again to produce further Compton electrons or photoelectrons.

In the third process, electron-positron pair production, matter is created out of pure energy. A gamma photon can interact with a nucleus to form simultaneously an electron and a positron, becoming annihilated in the process. The minimum amount of energy required for this process is the amount of energy represented by the masses of the electron and the positron, which is 1.02 Mev for the pair, any excess going into kinetic energy. However, this process dominates only at higher energies as previously indicated. The elusive nature of the positron (it was not discovered until 1932) is due to the fact that it readily undergoes a converse process, combining with an electron, to give off 1.02 Mev of radiation in the form of one or two photons, the pair disappearing in the process.

Penetration of Radiation

The preceding discussion indicated that the various interactions of the different radiations with matter were principally with the electrons and not the nuclei. Further, some of the interactions depend to a degree on the details of the binding forces between the electrons and nuclei of the target material, as in the photoelectric effect and the production of excited rather than ionized atoms by incident fast electrons or atomic radiations. However, for the Compton effect this binding energy is entirely negligible. In the process of forming a molecule from two or more atoms the forces acting on the electrons are somewhat changed, but this change is entirely negligible in considering the penetration of radiation.

To a very good approximation the penetrability of any material depends only on the numbers and kinds of atoms present, irrespective of the state of chemical combination. Also, the penetrability is highly independent of the physical state of the material. The range of a particular type of radiation in a solid, liquid, and gaseous form of a particular material will differ only inasmuch as the density of the material changes in these forms. A somewhat cruder but quite useful approximation is simply to compare different materials according to the number of electrons per unit volume they contain.

It was indicated earlier that atomic radiations transfer their energy in comparatively small units. At 1.0 Mev alpha particle or proton may undergo some tens of thousands of energy dissipating collisions before being stopped, and none of these collisions produces any appreciable deflection in the path of the charged particle. These circumstances produce a well defined penetration length or range, characterized by only a very slight statistical variation or range straggling. This range depends on the energy and increases somewhat more than proportionally. As the speed of the particle increases, the probability of transferring energy in an encounter decreases slightly. In the case of electrons, similar considerations apply with the exception that the frequent deflections experienced by electrons in the course of collisions introduce rather large discrepancies between the length of path traveled by a given electron and the net penetration range which has been reached at the end of the path. If the electrons were not deflected, they would have an equally well defined range. The net result of the deflections is that of a large number of incident electrons only very few reach this range and various fractions are stopped more or less short of this distance. This then corresponds to a maximum range with a considerably larger amount of straggling. For both types of radiation it is useful to summarize their penetration powers in the form of range-energy curves or range-energy equations for typical stopping materials.

The penetration of gamma radiation cannot be described by a range-energy relation. The disappearance of a gamma photon may take place in one act, as in the photoelectric or the pair-creation processes, or in a small succession of acts such as several Compton collisions followed finally by ejection of a photoelectron. This circumstance leads to quite different statistical consequences than was the case above. In effect the problem is—what is the probability that the gamma photon can penetrate a given distance without having undergone the single fatal collision or the small number of cumulatively fatal collisions? For the single collision the answer is that this probability decays exponentially. This is in accord with experience on the attenuation of visible light by an absorber such as a colored solution (Beer's Law) in which any given photon is either unaffected or totally absorbed in one act. In the case of a small number of collisions, the probability at first decays somewhat less than exponentially but gradually approaches an exponential decay. The extent of deviation depends on the energy of the photon, the nature of the stopping material and the distance of penetration under consideration. This deviation from exponential decay is graphically called the

5840
LIBRARY

"buildup factor." It must always be taken into consideration in design of shielding against high energy electromagnetic radiation. Overlooking of this "buildup" effect can lead to serious underestimates of shielding requirements. Quantitative calculations and nomographs taking this effect into consideration are available (Chappell 1958).

Induced Radioactivity

The processes discussed in the preceding sections represent the principal mechanisms whereby the energy of incident radiation is transferred to the target material. As the energy of the radiation rises above about ten Mev other processes can occur which, although they constitute only a very small part of the energy transfer, must be considered in practical applications of high energy radiation. These processes are the interaction of high energy radiation with the nuclei of the target material, leading to a variety of nuclear transformations which can induce radioactivity in the target.

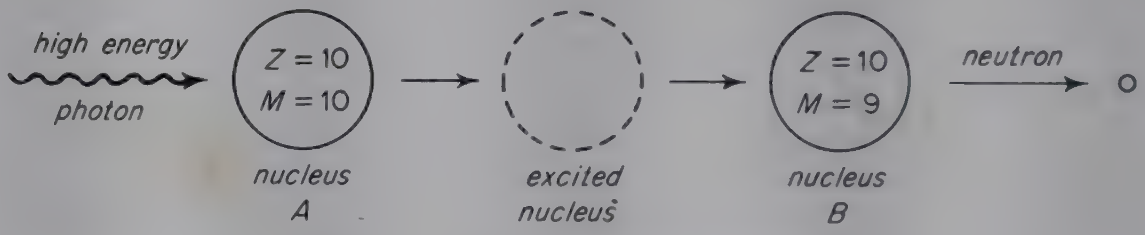


FIG. 13. NUCLEAR PHOTO EFFECT

Two types of processes are of particular interest, the ejection of particles from a nucleus by high energy gamma photons and the excitation of a nucleus by high energy electrons, followed by disintegration of the nucleus. The first process is the nuclear analogue of the photoelectric effect, differing in regard to the energy scale and the variety of particles that may be ejected. Prominent in this type of process is the ejection of a single neutron or proton and of lesser importance the ejection of several of these or of an alpha particle. The minimum energy requirement is set by the binding energy of the particle(s) and generally lies above ten Mev for the single particle process. The minimum energy requirements for ejection by high energy electrons are the same. However, the probability of the latter type of process is considerably smaller than that of the nuclear photo effect. In the case of high energy electron bombardment, it was mentioned earlier that at high energies a considerable fraction of the electron energy can be converted into high energy photons by the bremsstrahlung process. These photons may subsequently produce a nuclear photoeffect (see Fig. 13).

A further consequence of these primary nuclear effects is the occurrence of secondary nuclear reactions between the ejected particles and other target nuclei. The most important of these is the capture of the ejected neutrons by other nuclei, leading to the formation of other nuclear species which may be radioactive. It is to be noted that the nuclei which have undergone transformation by the photoeffect or electron excitation, may, on ejection of the particles, themselves become unstable radioactive species.

These processes have been studied in a variety of materials, including carbon, oxygen, nitrogen, sodium, and zinc, which represent species of biological importance. The primary transformations found are shown in Table 10 along with the activity and half life of the residual nucleus. There are some possible secondary reactions that may result from the capture of the ejected neutrons.

Table 10

THRESHOLD ENERGY LEVELS FOR THE PRODUCTION OF INDUCED RADIOACTIVITY IN VARIOUS ELEMENTS BY IRRADIATION WITH HIGH ENERGY ELECTROMAGNETIC RADIATION—THE ELEMENTS ARE LISTED IN GENERAL ORDER OF THEIR ABUNDANCE IN BIOLOGICAL MATERIALS[1]

Element	Type of Nuclear Change	Threshold Energy, Mev	Half-life of Product	Activity
C^{12}	gamma, neutron	18.7	21 min.	gamma[2]
O^{16}	" , "	16.3	2.1 min.	gamma[2]
N^{14}	" , "	10.6	10 min.	gamma[2]
P^{31}	" , "	12.4	2.5 min.	gamma[2]
K^{39}	" , "	13.2	1 sec.	gamma[2]
S^{32}	" , "	14.8	3.2 sec.	gamma[2]
Ca^{40}	" , "	15.9	1 sec.	gamma[2]
Fe^{54}	" , "	13.8	8.9 min.	gamma[2]
Mg^{24}	" , "	16.2	11.6 sec.	gamma[2]
Mg^{25}	" , proton	11.5	14.8 hr.	beta
Mg^{26}	" , proton	14.0	62 sec.	beta
Cu^{63}	" , neutron	10.9	10 min.	gamma[2]
I^{127}	" , "	9.3	13 days	gamma, beta
Al^{27}	" , "	14.0	7 sec.	gamma[2]
Be^{9}	" , "	1.7	very short	alpha
H^{2}	" , "	2.2	—	—
Na^{23}	" , "	2.6	2.6 yrs.	gamma[2]

[1] Adapted from Hannan (1956).

[2] Positron-electron annihilation radiation, 0.51 Mev.

Experimental studies, and supporting theoretical estimates, indicate that the amount of radioactivity induced in such samples by irradiation with 24 Mev electrons or photons is well below human tolerance levels for radiation doses which are high enough to produce useful radiation effects, such as sterilization of foodstuffs. For example, a dose of 5 megarads of 24 Mev electrons applied to beef would produce about 10^{-9} curies of sodium-22 per gram of beef. This radioisotope is a gamma emitter with a 2.6 year half life. For a beef consumption of 170 pounds per year, an individual would ingest about one ten-thousandth of the recommended maximum permissible amount of this isotope. Other long lived isotopes exhibit similar relations. Among the isotopes there are also some short lived ones which are produced in microcurie amounts. While these amounts themselves represent less than the permissible ingestion amounts, their short half-lives, of the order of minutes, are such that the safety factors will be tremendously enhanced during the inevitable time lag between processing and consumption. Clearly, these factors must be carefully considered in any particular processing application, but the above results suggest that problems of induced activity need not be a barrier to the employment of radiation in this very high energy range. For radiations of low energy, such as cobalt-60, which emits gamma photons of about 1.3 Mev energy, the induced activity problem simply does not exist, since the energy is too low for initiation of any of the above nuclear transformations.

Spatial Distribution of Ionization and Excitation

The spatial distribution of the ionization and excitation produced by a single radiation particle or photon is different in the three types of radiation considered.

In the case of alpha particles and atomic radiations the path of the ionizing particle itself is straight. The rate at which energy is transferred to the target material is high enough that ionizations and excitations occur continuously along the path of the particle, tending to be somewhat more dense near the end of the particle track because the rate of energy transfer increases as the particle is slowed down. The secondary electrons ejected by these particles are of quite low energy, not exceeding about 2 kv. for a 4 Mev alpha particle. Those electrons which have this maximum energy are ejected in a forward direction; as the ejection energy is decreased the direction of ejection gradually changes to perpendicular to the track. The range of these perpendicular electrons is, of course, governed by their energy, in this case both are very small. These electrons may travel roughly ten molecular diameters before they themselves have lost their energy by further excitation and ionization. The secondary electrons are often termed delta-rays and their tracks are termed "spurs."

In the case of high energy electrons, because of occasional large-angle deflection, the tracks themselves consist of an irregular sequence of straight line segments. In these segments the density of ionization and excitation is somewhat less than for atomic particles because of the higher velocity of the electrons. Again, as the end of the electron path is approached the density rises. In contrast to atomic particles, the electrons can eject secondary electrons which are for the most part of low energy, but may have appreciable energy. The low energy group of electrons again forms spurs off the primary track but the small number of high energy electrons form distinct tracks of their own, with associated spurs.

The energy dissipation of a gamma photon occurs in a small number of discrete steps each leading to the formation of high energy electrons. Thus the "track" of a gamma photon consists of a number of more or less separated high energy electron tracks with all their features (see Fig. 14).

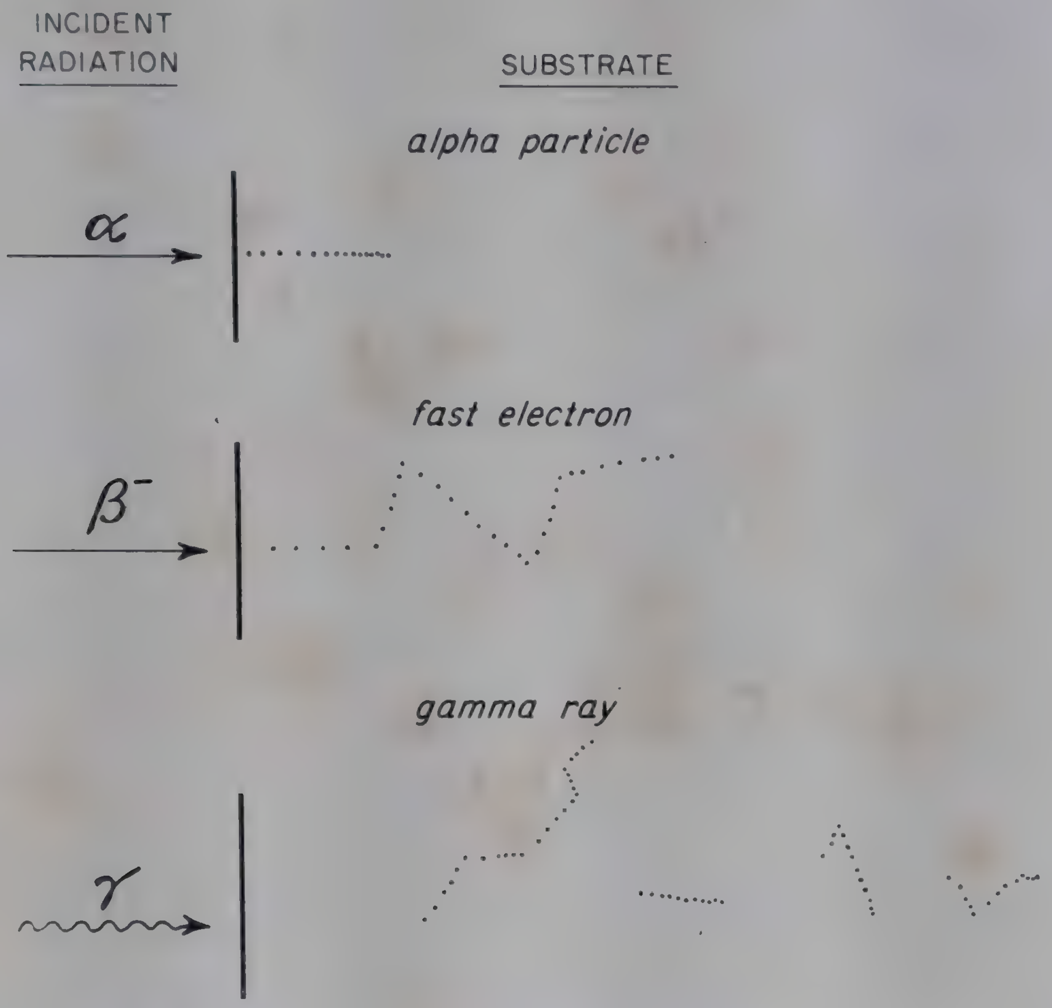


Fig. 14. Schematic Diagram of Tracks of Ionizing Particles

The situations described above are essentially similar in liquids of non-metallic solids, and differ only in the expanded scale of distances in a gas. The three cases are well illustrated in the accompanying cloud chamber diagrams in which the individual ionizations are made visible by the fact that in the cloud chamber each ionization forms the nucleus for the growth of a liquid droplet.

Precise numerical descriptions of the distribution of excited and ionized molecules are difficult to make but a reasonable estimate is that in liquid water, for example, a typical spur may be roughly twenty angstroms in length, four thick, and may contain of the order of five excited and ionized molecules. It is evident from this that the concentration of excited and ionized species is exceedingly non-uniform and, in the spurs themselves, approaches fantastic values when expressed in ordinary concentration units. If one were to apply the usual thermodynamic reasoning to compute the temperatures necessary to obtain such concentrations of activated species, the resulting temperatures are in the thousands of degrees.

These considerations provide the clue to the unusual capability of radiations to induce chemical and biological change. Although a material exposed to very high intensities of radiation may show an only barely perceptible temperature rise, this rise is brought about by a time and space averaging of the temperature of the preponderant part of the material which is completely unaffected by radiation together with the initial "temperature" of those very small regions of the material in which the radiation energy is transferred. It is in these regions that chemical and biological changes are initiated. The non-uniform "temperature" and "concentration" brought about by the passage of radiation starts to equalize immediately after its formation. Of particular importance in this connection is the radiation-induced ionization. Not only will the new chemical species tend with time towards a uniform concentration but the charges separated by the radiation impact will recombine. The detailed unfolding of this complex sequence of events is dependent on the rigidity of the material, the dielectric properties of the material and the charge, molecular configuration and chemical reactivities of the various species and the bulk material.

BIBLIOGRAPHY

Bethe, H. A., and Ashkin, J. 1953. Passage of Radiations Through Matter. Experimental Nuclear Physics. Editor, E. Segre. Vol. I. John Wiley and Sons, Inc., New York.

Bohr, N. 1948. The Penetration of Atomic Particles Through Matter. Det. Kgl. Danske Videnskabernes Selskab. Mathematisk-fysiske Meddelelser XVIII, Denmark.

Chappell, D. G. 1958. Gamma ray attenuation with buildup in water. Nucleonics *16,* No. 7, 80–84.

Evans, R. D. 1955. The Atomic Nucleus. McGraw-Hill Book Co., Inc., New York.

Freiling, E. 1956. Nomogram for Radioactivity Induced in Irradiation. Nucleonics *14,* No. 8, 65–71.

Glass, R. A. 1958. Radioactivities produced in foods by high energy electrons. QM. Food and Container Inst. Research Report, S-572, No. 3, Chicago, Illinois.

HANNAN, R. S. 1956. Science and Technology of Food Preservation by Ionizing Radiation. Chemical Publishing Co., New York.

HERSCHMAN, A. 1956. Induced radioactivity considerations in food. QM. Food and Container Inst. Research Project Report, Proj. 7-84-01-002, File No. 0203AO. Chicago, Illinois.

MAGEE, J. L. 1953. Radiation Chemistry. Ann. Rev. Nuclear Science *3*, 171–192.

MASSEY, H. S. W., and BURHOP, E. H. S. 1952. Electronic and Ionic Impact Phenomena. Oxford University Press, London, England.

RICHTMYER, F. K., and KENNARD, E. H. 1947. Introduction to Modern Physics. Fourth Ed. McGraw-Hill Book Co., Inc., New York.

PART II

Radiation Source Technology

CHAPTER 6

Radiation Sources

The discovery of nuclear fission uncovered the possibilities of converting a portion of the mass of certain elements into energy; the fission of the uranium-235 in a pound of natural uranium yields the energy equivalent of 22,000 pounds of coal plus useful by-products. With the subsequent establishment of the fact that a chain of fission events could be initiated and controlled came its application in the form of a nuclear reactor. Both the energy released as electromagnetic and particle radiation, and that contained in the radioisotopes are useful.

Paralleling the developments in reactor technology, a variety of electromechanical devices have been developed which generate high energy radiations with intensities sufficient to be important in industrial applications; these include the Van de Graaff generators, linear accelerators, capacitrons, and others. Both approaches are valuable means of developing radiation.

Nature of Fission Reaction

The fission of uranium was first experimentally demonstrated by Hahn and Strassmann in 1939 and correctly interpreted by Meitner in the same year. It was subsequently found that by capturing a neutron the uranium nucleus underwent fission and yielded two fragments of approximately half the mass of the original nucleus, accompanied by the release of several neutrons and considerable amounts of energy. An example of this type of reaction is shown in the following equation:

uranium-235 + neutron →
element mass nos. 95 and 139 + 2 neutrons + energy

The large amount of energy released in fission is the result of the direct conversion of mass into energy, predicted by Einstein in 1905.

In the conversion of a nucleus of mass 235 and one neutron, into the two fission fragments of mass numbers 95 and 139 and the accompanying two fission neutrons it is found that a certain amount of mass has in fact disappeared. The precise mass of the starting materials amounts to the total of 236.127 mass units (235.118 + 1.009) while that of the products of the reaction total 235.904 mass units (94.936 + 138.950 + 2.018). The difference in mass amounting to 0.223 mass unit has been converted into energy equivalent to 200 Mev. This energy is liberated in the form of radiation and kinetic energy of the product fragments.

The actual mass distribution of fission fragments was shown in Fig. 5. In Table 11 is shown the distribution of the energy released in fission as apportioned among the neutrons, gammas, fission fragments, beta rays, and neutrinos. It is to be noted that about 80 per cent of the total energy is released in the form of kinetic energy of the fission fragments. It has been mentioned earlier that the range of these fission fragments is quite short; the energy is thus nearly completely dissipated into heat in the neighborhood of the site of the fission event.

Nature of Chain Reaction

The experimental fact that at least one neutron is released in the fission process opens the possibility of a self sustaining nuclear chain reaction, and the possibility of a nuclear explosion. If more than one of the released neutrons is captured by other uranium nuclei resulting in fission the process can propagate itself and can in fact become explosive. For example if two neutrons released in a single fission can yield two additional fission reactions, and so on, it is readily seen that at each successive step the number of fission events doubles, and in a fraction of a second the process can become violent enough to produce an explosion.

In practice the problem of obtaining and sustaining a controlled chain reaction is one of exceedingly great complexity (Weinberg and Wigner 1958). Although two neutrons may be released in a fission event, it does not necessarily follow that both will be captured by other uranium nuclei. The neutrons may be captured by other nuclei which do not undergo fission as a result of neutron capture.

TABLE 11

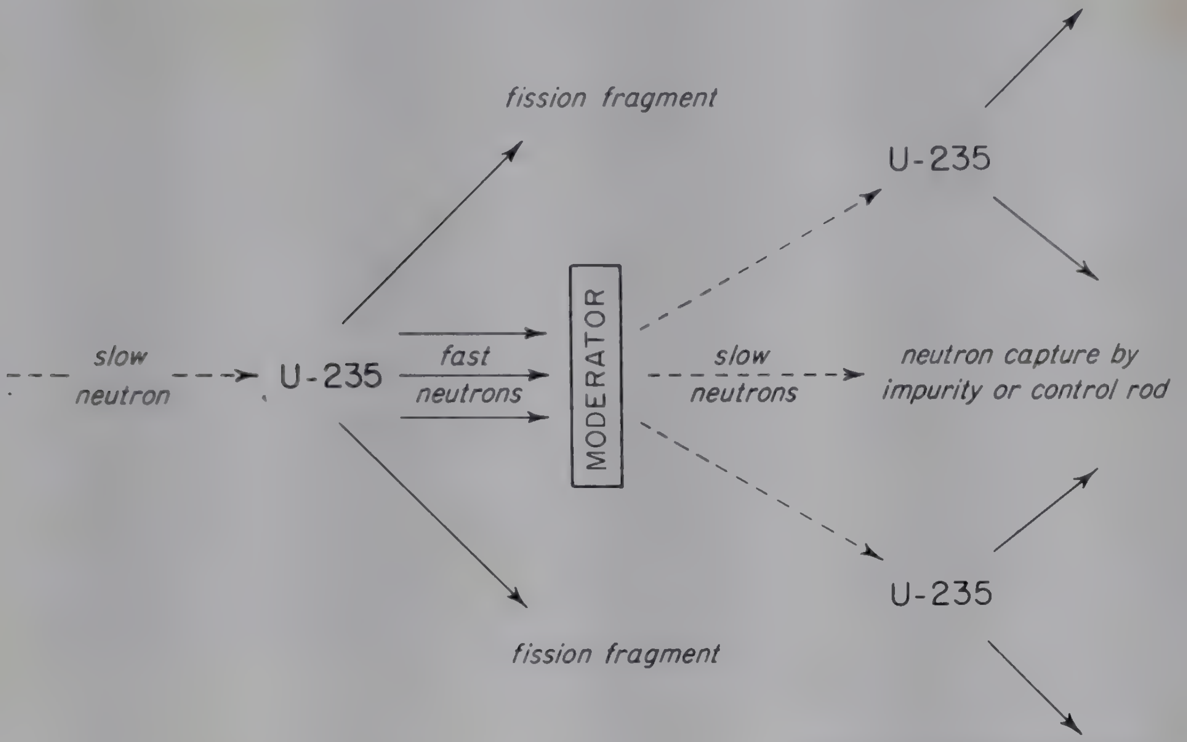
DISTRIBUTION OF FISSION ENERGY[1]

	Mev
Kinetic energy of fission fragments	168
Fission neutron energy	5
Instantaneous gamma energy	5
Beta particles from fission products	6
Gamma rays from fission products	6
Neutrinos from fission products	10
	200

[1] From Glasstone (1958).

Further, depending on the spatial arrangement of the uranium nuclei, some of the neutrons may completely escape from the assembly of uranium in which it is intended to produce a chain reaction.

In Chapter 2, the energy of a thermal molecule was seen to be only a few hundredths of an electron volt. The neutrons obtained from the fission reaction are much more energetic (Table 11). Because it has been found that the neutron capturing by uranium is optimum in the thermal range, actually less than one electron volt, a problem then develops in that if the neutrons are not slowed, they escape, and are useless in maintaining the fission chain. The solution is found in making these "fast" neutrons collide with nuclei of a mass nearly that of the neutron itself. At equal mass there is a maximum of energy transfer, i.e., slowing down. On the basis of mass, hydrogen would be ideal; however it is not found functional. Water, certain organic molecules and beryllium have been found useful. A compromise must be made between the weight of the nuclei and the density of the material. These substances when employed to slow down neutrons are called moderators. The scheme of events is shown in Fig. 15.



Courtesy of Phillips Petroleum Co.

FIG. 15. SEQUENCE OF EVENTS IN THERMAL REACTOR CORE

The relative abundance of fission fragments was shown in Fig. 3.

Another problem arises. Real materials have impurities with intrinsic capabilities of capturing neutrons in an unproductive fashion. So, in fact, a real reactor, composed of real materials must be a compromise with real amounts of impurities, and the fission process itself continuously produces a variety of fission fragments which in fact are then impurities.

It also follows in practice that some neutrons will be produced at or near the surface of the "core" and will escape. Materials have been found which can be placed around the core and serve to reflect many of the escaping neutrons. Essentially the reflector components have the same general properties as moderators. The reflector is functional also in shielding personnel who must work near the reactor. It is to be noted that reflectors, due to their intrinsic nature as good neutron shields are inefficient gamma ray barriers.

Control of a reactor once the chain of fission events begins is accomplished by restricting the neutron flux in the core. This flux is controlled by the use of rods of cadmium or boron steel. Such rods inserted into the core absorb excess neutrons, and keep the multiplication factor to near one. If the rate of fission reaction becomes excessive, the system breaks apart rather than explodes, due to the inability to remove the tremendous quantities of heat generated internally. Each piece being less than critical in size, the chain reaction ceases, and no explosion occurs. Radiation and contamination dangers are substantial however. The use of control rods is then seen as a control of the neutron flux in the core, and adjustment of the rods inward to stop or outward to start the reaction is all that is required. Readjustment of the rods is then required to obtain desired results.

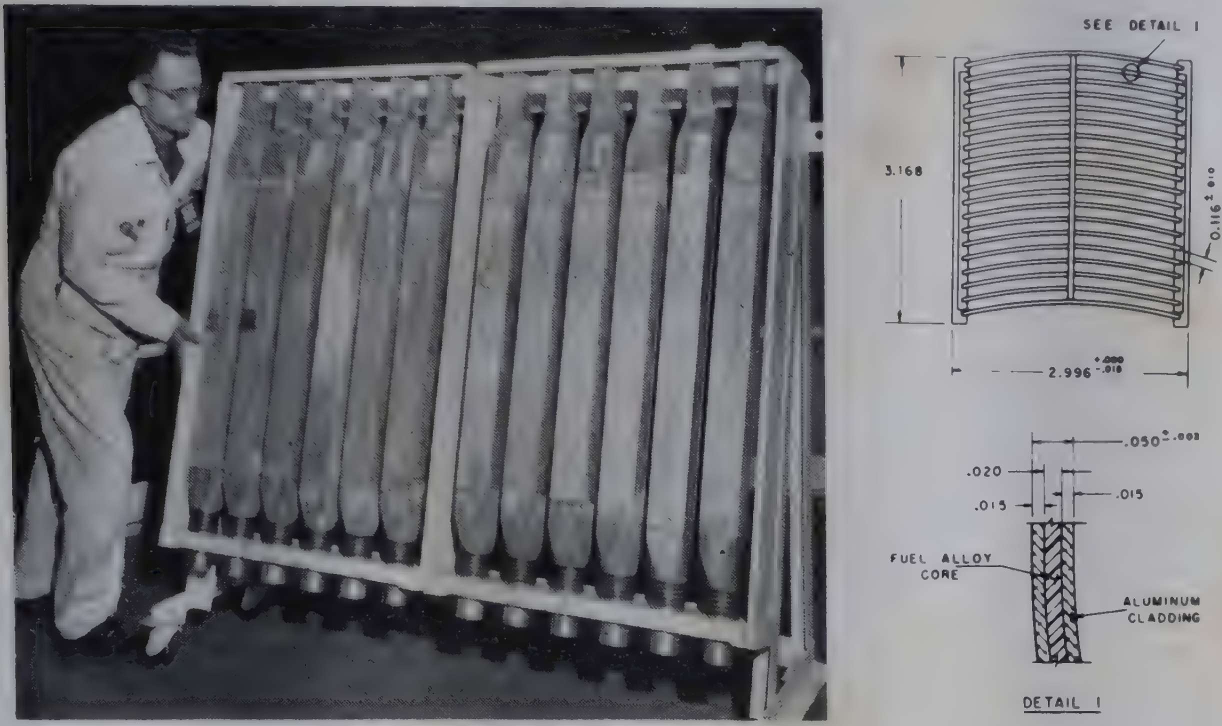
The critical size of a system of fissionable components is one in which the number of neutrons produced by fission equals or is slightly larger than the losses of neutrons. If smaller than critical, the term used is subcritical. A supercritical size is one greater than the minimum critical size. The critical size is variable and dependent upon the fissionable material used, the relation to the moderator, the shape and organization of the components, and the presence of neutron capturing substances.

While there are perhaps thousands of methods of solving these problems in reactor engineering, some solutions are adapted to yield reactors whose main product would be high neutron fluxes for research purposes, some reactor designs yield compactness, useful for submarine and airplane propulsion, some reactors are designed to breed fissionable materials, others are designed to yield energy for use in power production.

No matter what the design, all have one problem in common—disposal of radioactive wastes from the reactors. These radiation sources can either be dumped in the ocean or buried in the ground in non-productive use, or they can be put to work as radiation sources. Non-productive disposal is less desirable than employment in radiation applications. There are perhaps hundreds of millions of curies in such non-productive use currently, with larger amounts to follow, which might be usefully employed.

Reactor Types

Nuclear reactors are grouped into classes according to the neutron energy region in which nuclear fission occurs—thermal (slow), fast, and intermediate. *Thermal* reactors operate with slow neutrons. Because the neutrons released in the fission process are very energetic, it is necessary to retard them, and this may be accomplished by the presence of a moderator, characterized by having low atomic weight. It should not capture neutrons in significant degree. The practical moderator substances found are water, heavy water, carbon in the form of graphite, certain organic compounds, beryllium, and beryllium oxide. Heavy water (deuterium oxide) is the most efficient. Thermal reactors are flexible in the nature and properties of the components, mainly in fuel and moderators (Glasstone 1958).



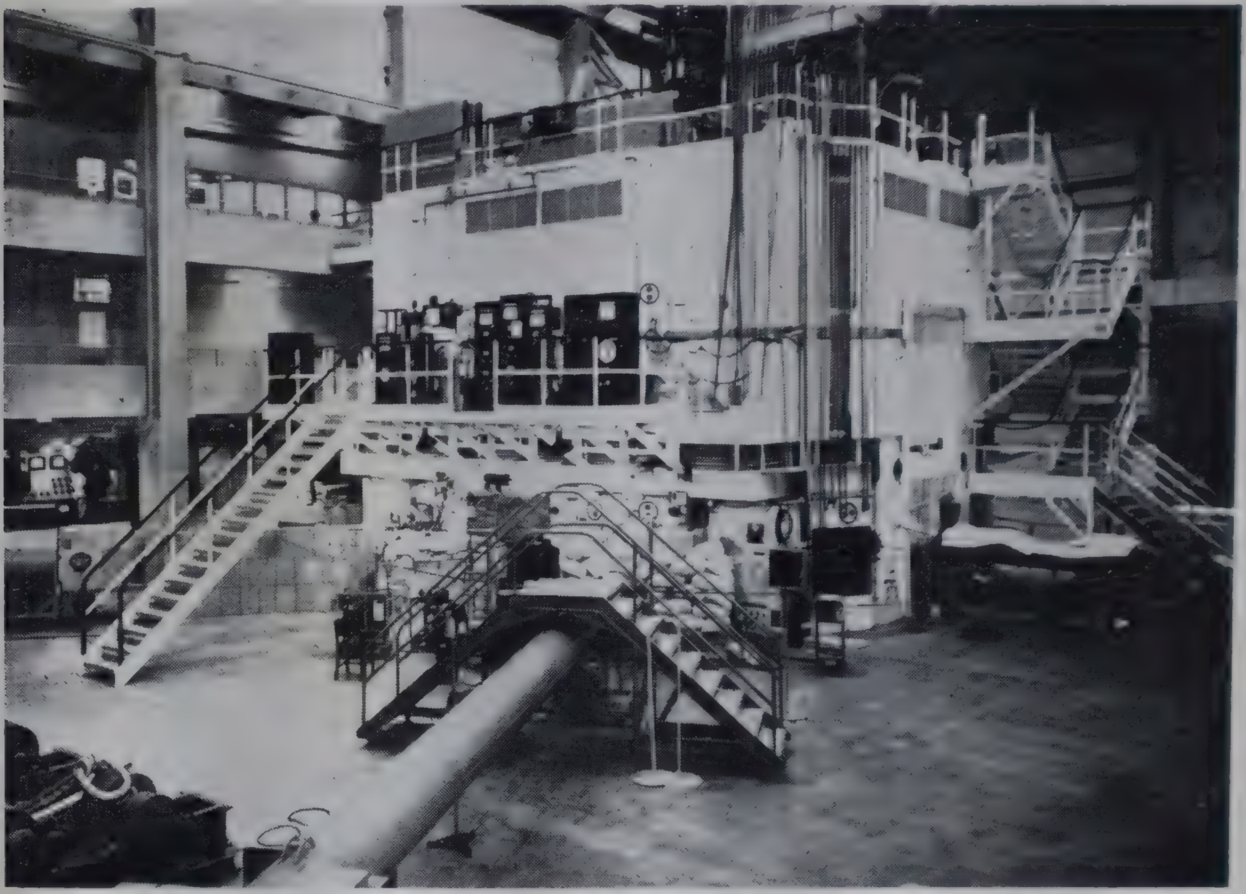
Courtesy of Phillips Petroleum Co.

FIG. 16. FUEL ASSEMBLY UNITS OF ENRICHED URANIUM FOR REACTORS

Ends of elements are open to allow coolant to pass freely from top to bottom

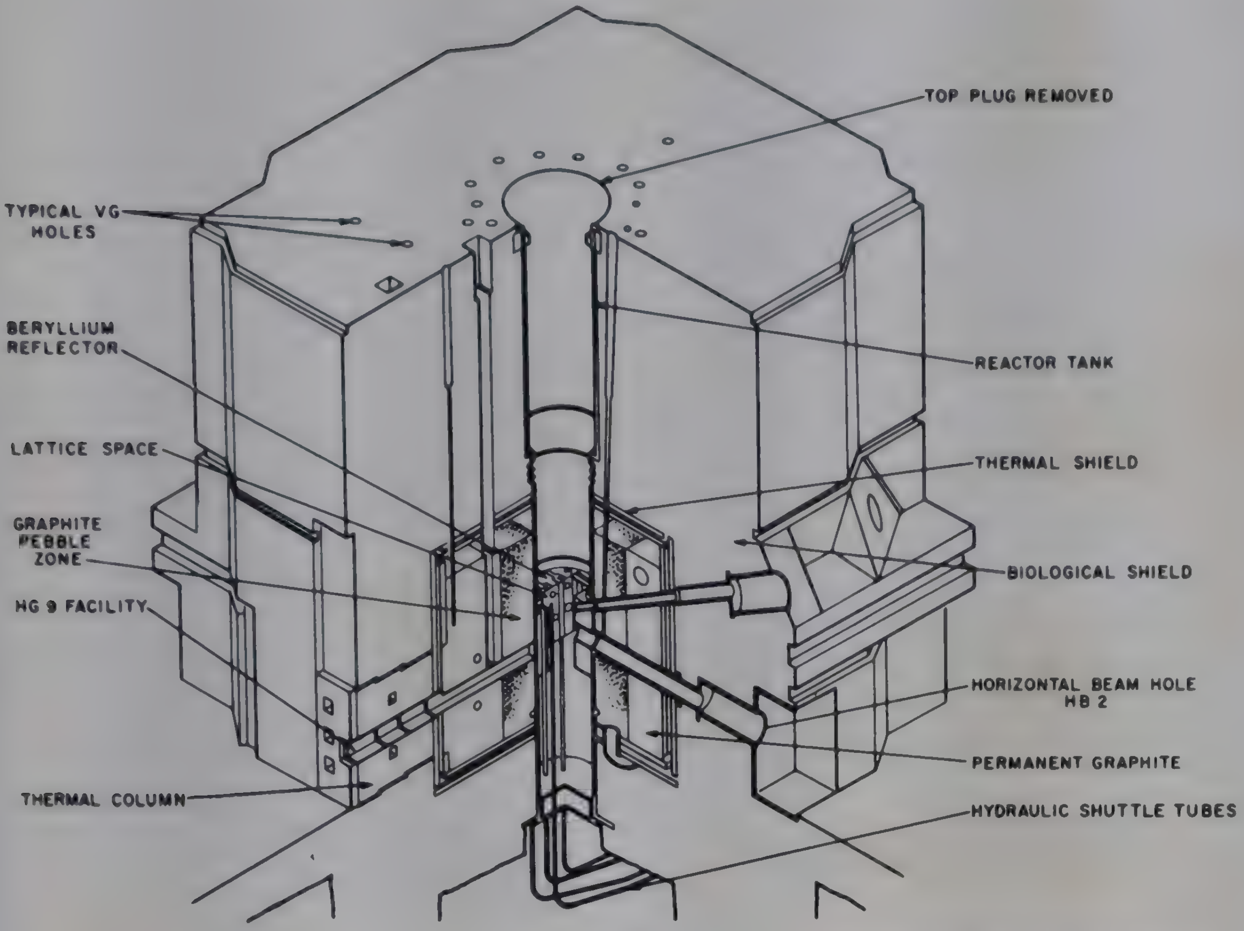
The series of events which occur in the core of a thermal reactor are shown in Fig. 15. The presence of fission fragments and other impurities eventually leads to their interference with the operation of the reactor due to the capture of neutrons. This fuel is then considered "spent," and is replaced. The uranium is separated and recovered from these spent fuel elements. The sequence is shown pictorially in Figs. 16, 17, and 19, for the Materials Testing Reactor at Arco, Idaho. A schematic diagram of this research reactor and core are shown in Figs. 18A and 18B.

A *fast* reactor operates with unimpeded neutrons, released in fission, called fast neutrons. The use of a moderating substance, whose main role is to slow the generated neutrons, is then undesirable. Because the general ability to interact with fast neutrons is small, the choice of construction materials for a fast reactor is greater than for the thermal reactors. There is a restriction however in the fuel requirement. Where the thermal type functions over a rather wide range of uranium, from natural to highly enriched, or with plutonium, the *fast* reactor usually requires enriched fuel containing upwards of 25 per cent of fissionable uranium-235 or plutonium-239. A fast reactor requires large amounts of such fissionable material to establish a chain of events, but the requirement of a moderator being absent, the critical size may be small, e.g., as small as a basketball. Naturally, heat removal from such a restricted volume is difficult. The main attraction of fast reactors is their ability to breed fissionable material, plutonium-239 or uranium-235. As long as the neutrons released are in close balance with the requirement for chain reaction, below the prompt critical value, the *fast* reactor is controllable with no greater difficulty than the thermal reactor.



Courtesy of Phillips Petroleum Co.

FIG. 17. MATERIALS TESTING REACTOR, ARCO, IDAHO
Fuel elements positioned as shown in Fig. 18B


TOP PLUG REMOVED
TYPICAL VG HOLES
BERYLLIUM REFLECTOR
REACTOR TANK
LATTICE SPACE
THERMAL SHIELD
GRAPHITE PEBBLE ZONE
BIOLOGICAL SHIELD
HG 9 FACILITY
HORIZONTAL BEAM HOLE HB 2
PERMANENT GRAPHITE
THERMAL COLUMN
HYDRAULIC SHUTTLE TUBES

Courtesy of Phillips Petroleum Co.

FIG. 18A. SCHEMATIC DIAGRAM OF MATERIALS TESTING REACTOR

Between *thermal* and *fast* reactors is another category which operates in the intermediate range of neutron energies. Moderators are used in smaller amounts than in a thermal reactor. While such reactors can be designed to breed plutonium-239, they have been found less effective than fast types, and hence are of less interest.

Reactor Power

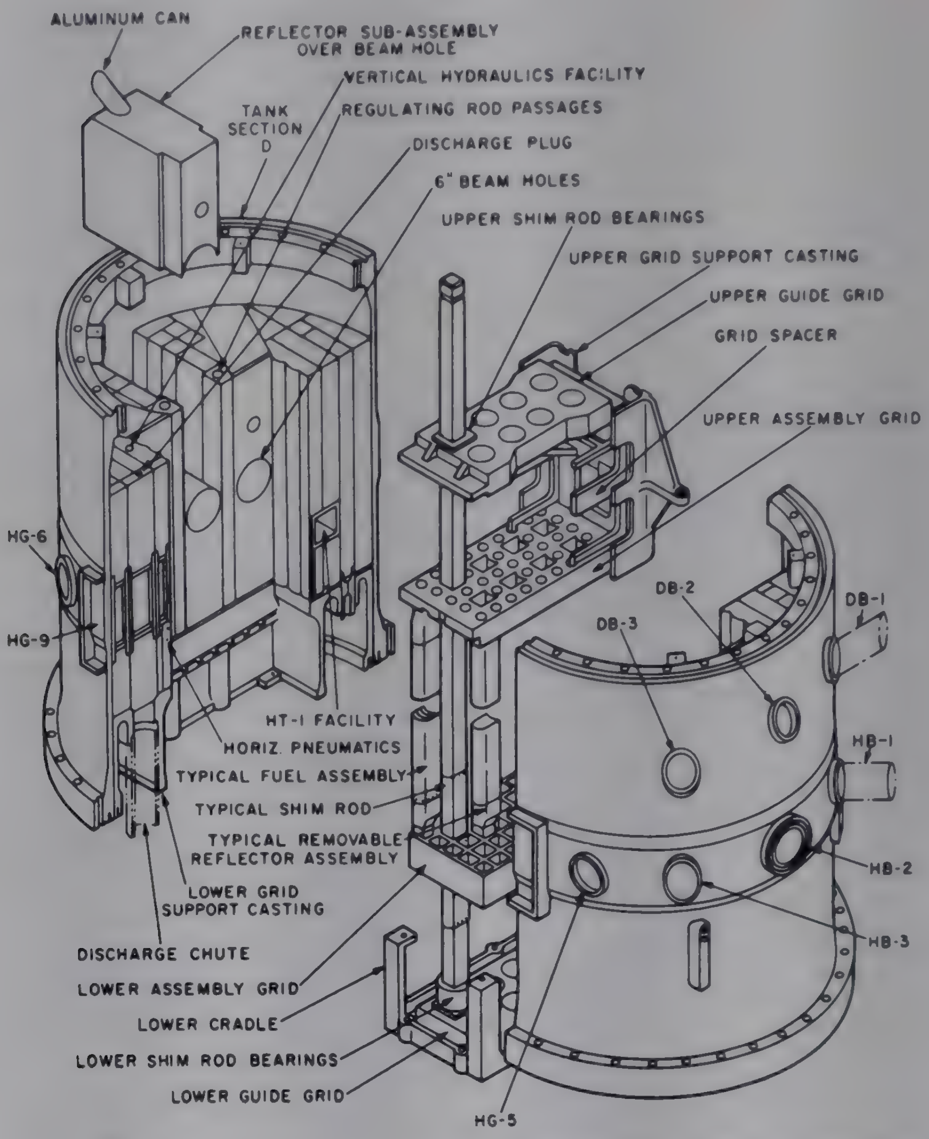
One watt of power is obtained from 3.1×10^{10} fissions per sec.

The power obtained from a reactor measured in watts is given in the following equation:

$$P\ (\text{watts}) = 2.5 \times 10^{-8}\, w\, n\, v$$

where $w =$ weight of uranium-235 in pounds
$n =$ average neutron density per cm.^3
$v =$ average-speed of neutrons in cm. per sec.

The quantity $n\,v$ is the neutron flux, and is measured in neutrons per sq. cm. per sec. The neutron flux is equal to the sum of the distances moved by all the neutrons in one cm.^3 in one sec. and is assumed to be uniformly distributed.


ALUMINUM CAN
REFLECTOR SUB-ASSEMBLY OVER BEAM HOLE
VERTICAL HYDRAULICS FACILITY
TANK SECTION D
REGULATING ROD PASSAGES
DISCHARGE PLUG
6" BEAM HOLES
UPPER SHIM ROD BEARINGS
UPPER GRID SUPPORT CASTING
UPPER GUIDE GRID
GRID SPACER
UPPER ASSEMBLY GRID
HG-6
HG-9
DB-2
DB-3
DB-1
HT-1 FACILITY
HORIZ. PNEUMATICS
TYPICAL FUEL ASSEMBLY
TYPICAL SHIM ROD
TYPICAL REMOVABLE REFLECTOR ASSEMBLY
HB-1
HB-2
HB-3
LOWER GRID SUPPORT CASTING
DISCHARGE CHUTE
LOWER ASSEMBLY GRID
LOWER CRADLE
LOWER SHIM ROD BEARINGS
LOWER GUIDE GRID
HG-5

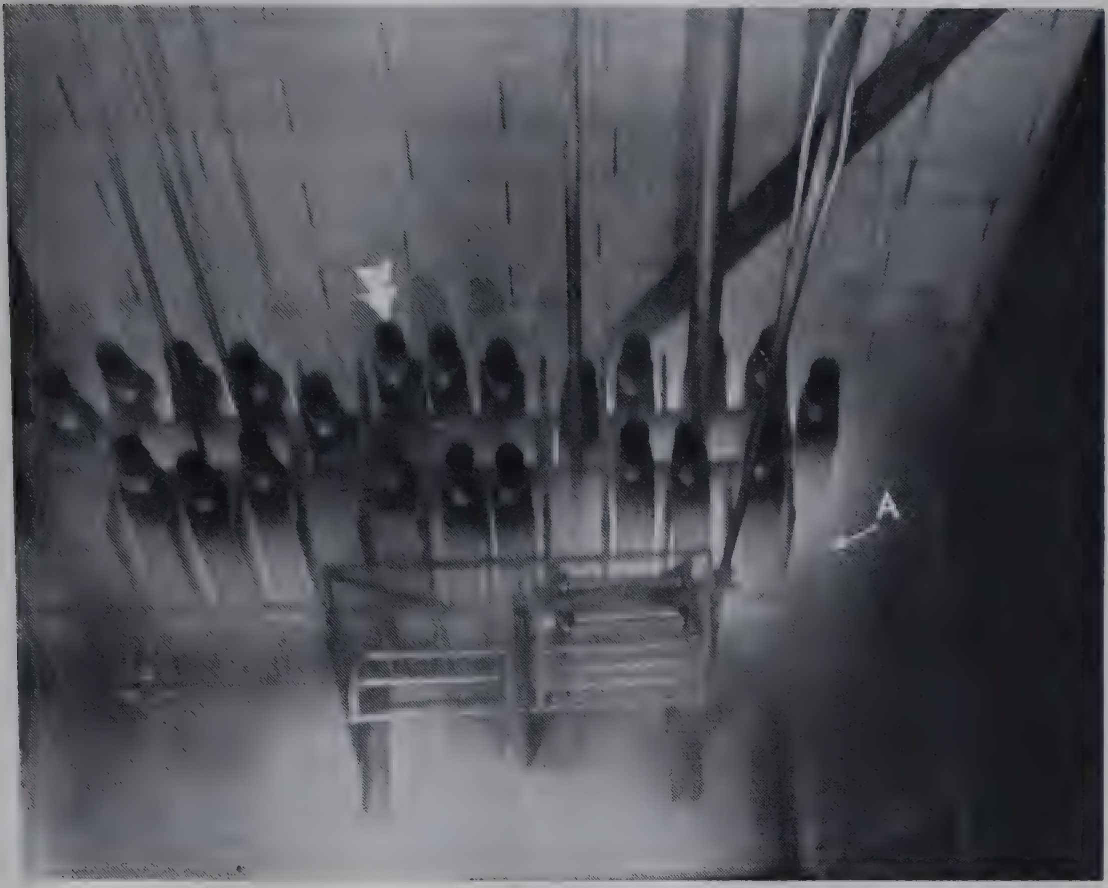
Courtesy of Phillips Petroleum Co.

FIG. 18B. SCHEMATIC DIAGRAM OF MATERIALS TESTING REACTOR

Reactors are classed on the basis of their use, such as the research reactor, the production reactor, and power reactors.

The earliest research reactors used natural uranium and graphite moderators with or without forced air cooling. These reactors are usually large, requiring some 30 tons of uranium and 300 tons of graphite. Another type is called the "water boiler." This is of the homogeneous type, having the fuel dissolved in water. The mixture is contained in a stainless steel sphere or cylinder. The first of these in the U. S. was developed at the Los Alamos Scientific Laboratory and used two pounds of U-235 and 3.75 gallons of water. The core was contained in a graphite reflector some 4.5 feet thick. Heat was removed by passing cold water through the core. It should be noted that the water need not boil.

The "swimming pool" reactor usually consists of fuel elements of the sandwich plate type. At least six pounds of U-235 are required. The



Courtesy of U.S. Atomic Energy Commission

FIG. 19. A URANIUM-235 REACTOR

After about 25 per cent of the uranium in the assembly has undergone fission in the reactor, spent fuel element is ejected into water canal. Units now intensely radioactive—Cerenkov radiation seen as glow. Photo taken above about 14 feet of water. These elements are useful in radiation technology. Each unit is in the several hundred thousand curie range of activity

fuel elements are suspended vertically near the bottom of a pool of water, some 20 feet deep. Water acts as moderator, reflector, and shield. Cooling may be by convection, but with heat exchanger equipment external to the reactor, to increase the cooling effect, operation near 1000 kilowatts is possible. The Materials Testing Reactor (shown in Figs. 16–19) is of this type. This reactor was designed to yield high neutron flux to test reactor component materials. The fuel elements used in the MTR are shown in Fig. 16. A heavy water (D_2O) moderated reactor of the same general type is operating at the Argonne National Laboratory.

The Los Alamos Fast Reactor uses plutonium-239 as fuel, and, as the name "*fast*" implies, has no moderator. The reflector was uranium metal and steel, and the coolant mercury. Mercury is not used in a thermal reactor as a coolant due to its great capture of slow neutrons; fast neutron capture is negligible.

Production reactors have been designed to yield fissionable material as a fuel for other reactors, and for weapons. Some dual purpose reactors have been proposed in which fissionable material and power would be products. The Calder Hall reactors in England are dual purpose. The use of production reactors to manufacture radioisotopes is worthy of note. Here a stable element is exposed to neutrons in a reactor, resulting in n,γ reactions, a radioisotope of the stable elements introduced into the reactor being obtained. The graphite moderated reactors at the Oak Ridge and Brookhaven National Laboratories are used in part for radioisotope production. Radioisotopes have found widespread use as a powerful new research tool, and industrial applications of great significance are appearing.

A power reactor is designed to convert the heat obtained from the fission process into electrical energy. The potential of one pound of uranium-235 or plutonium-239 to yield the heat energy equivalent to that obtained from burning 3,000,000 pounds of coal is challenging to an energy hungry world. The steam to drive turbines is produced either directly or through a heat exchanger system.

Because the maximum operating power of a reactor is determined by the rate at which the core can be cooled, a limiting factor is the ability to remove heat. If neutron fluxes are high, hence fission high, there is danger in thermal reactors of building up the fission fragment gaseous xenon-135, a reactor poison, which interferes with its operation. Fast reactors do not experience this difficulty.

The "pressurized water reactor" is a heterogeneous type using high pressure (2,000 lbs. per sq. in.) water as moderator-coolant, which is circulated into the reactor, through the core, and out into a heat exchanger system. This was the system used in the USS *Nautilus,* the first nuclear powered vehicle or vessel. It uses highly enriched uranium-235 with water as the moderator. The core size is small of necessity. Where space is no limitation, less enriched fuel can be used in a pressurized system. The Pressurized Water Reactor built at Shippingport, Pennsylvania, is the first full scaled nuclear power plant in the U. S. The reactor is designed to have a minimum electric power capacity of 60,000 kw., and it is being increased to 100,000 kw.

"Boiling water" reactors are used to take advantage of the efficiency in heat removal by converting water to steam; the water moderator-coolant is boiled directly in the core of the reactor. At the Argonne National Laboratory the Experimental Boiling Water Reactor generates steam at 600 lbs. per sq. in., and uses partially enriched fuel.

Homogeneous reactors may use aqueous solutions of uranyl sulfate as fuel similar to that of the "water boiler" type. The fuel is carried in the water which also acts as the moderator-coolant medium, and is passed externally to be cooled with a heat exchanger. While fuel elements in solid fuel reactors must be periodically replaced and after cooling must be reprocessed and refabricated, the homogeneous liquid fuel reactor has the advantage that it is possible to extract certain fission fragments and other impurities continuously during actual operation. The fuel solution is very corrosive, however.

Graphite moderated reactors use solid fuel elements with graphite as the moderator. Liquid sodium has been used as a coolant, which is passed from the core to a heat exchanger system for cooling. Such sodium loops are useful in irradiation technology, taking advantage of their gamma flux developed by the n,γ reaction.

Uranium-235 is the only fissionable material occurring in nature. Natural uranium contains about 140 times more U-238 than U-235. In a reactor, U-235 is used in the process of fission, yielding neutrons which initiate another fission event or are captured by U-238. This in turn eventually yields plutonium-239, also fissionable. A similar story applies to thorium-232, which yields U-233. It appears that the long term future of the nuclear energy industry will be dependent upon consuming plutonium-239 (or uranium-233) and at the same time regenerating them from U-238 and Th-232. Such a reactor using plutonium as the fuel undergoing fission yields neutrons a portion of which may be captured by U-238 which in turn generates plutonium-239. The process is called breeding, and has been made to occur for uranium-233 from thorium-232.

The "fast breeder" reactor with highly concentrated uranium-235 or plutonium-239 may use liquid sodium as a coolant. A breeding layer of uranium-238 serves effectively as the reflector. The heated sodium coolant is passed externally through a heat exchanger system.

It is inherent in the nature of a reactor that a particular unit designed for a specific purpose, e.g., power generation, may be utilized towards other ends as well. The breeder reactor is an excellent example. It is interesting to speculate on and important to investigate possible multiple purpose reactors which combine several functions such as power generation, fuel breeding, isotope production, and radiation processing.

Reactor Core as Radiation Source

A direct method of irradiation would consist of exposing material directly in the core of a reactor.

Because of the high neutron flux present in the core, substantial problems in induced radioactivity will result if a material is exposed directly in the core. The use of neutrons in the sterilization of food materials has been studied. It is found that after approximately 90 days there remains no significant induced radioactivity, but it appears that such a method is less attractive than irradiation by other means.

In addition to introducing the substrate to be irradiated internally in the reactor, it would appear that energy is available by using fission fragment recoil energy. From Table 11 it is seen that more than 80 per cent of the energy of fission is available as kinetic energy of the fission fragments. This is to be compared with the approximately 12 per cent availability of gamma and neutron energy. However the use of fission fragment recoil energy requires an intimate mixing to minimize self-absorption of the fragment energy in the fuel itself.

Since the range of fission fragments is very small, a fraction of a millimeter, in order to utilize effectively the energy available, it is necessary to construct fuel elements of exceedingly small dimensions. Harteck and Dondes have made the imaginative approach of using uranium fuel incorporated into extremely fine glass fibers. These fibers are so thin that an appreciable portion of the fission fragment energy is recoverable. Such an approach is well suited to the irradiation of gaseous and liquid substrates. However, here again serious problems of contamination of treated material may occur.

A number of reactors have been proposed for power production employing a coolant of liquid sodium. The sodium so incorporated undergoes the following reaction:

$$\mathrm{Na}^{23} + n \rightarrow \mathrm{Na}^{24} \rightarrow 2.75\ \mathrm{Mev}\ \gamma + 1.37\ \mathrm{Mev}\ \gamma + 1.39\ \mathrm{Mev}\ \beta + \mathrm{Mg}^{24}$$

The sodium-24 emits useful 1.37 and 2.75 Mev gamma rays with a half-life of approximately 15 hours. An external loop through which the sodium exits from the reactor could be passed through a radiation processing area, without disturbing the reactor operation. In fact, such a power reactor is being built near Lincoln, Nebraska, using sodium as a coolant. Sufficient quantities of radiation become available to sterilize nearly a million pounds of animal flesh a month as a by-product in the production of electric power. If power production expands in its use of the nuclear reaction as a source of energy, it is anticipated that such power plants may indeed be a major radiation service rendering agent of the next decade. Radiation services offered by such utilities could be an important factor in the development of radiation as an industrial process. Such dual purpose reactors will make both power and radiation services economically even more attractive.

Fission Products

A gram of U-235 yields approximately one gram of fission products and 24,000 kw. hr. of heat. As indicated above, the fission products are radioactive and are useful by-products from the reactor. Because of the growth of reactor applications, it would seem inevitable that fission products will be a major factor for consideration as radiation sources. However, while fission products will be available in increasing quantities, the cost of making these available for industry has not been completely evaluated. It would appear that fission products used for radiation sources may well be in the form of spent fuel elements or in gross, mixed, or separated radioisotopes.

Spent Fuel Elements

Fuel elements which have been exposed in a nuclear reactor for long periods of time at high specific power provide the highest intensity radiations available in quantity. The fuel elements coming closest to these requirements are those used in power plant reactors. Most of these fuel elements consist of rods or plates which must be allowed to cool prior to being reprocessed. During the period of cooling, high intensity radiation is available. Such fuel elements form the bulk of the radiation sources used in the United States in radiation technology, in particular those studies dealing with applications in the area of food and agriculture. Typical of such fuel elements are those shown in Fig. 19.

Gross Fission Products

Because spent fuel elements contain substantial usable fissionable material, the fuel elements are generally reprocessed chemically, leaving fission products, salts, contaminants, and water. This mixture with the water present is called the gross fission product. Initially the specific activity is near a curie per gram which is low, entailing long dwell times for the irradiated products. In addition, their rapid decay in a few months to a point of low intensity, involves costly transportation and replacement problems. The U. S. Atomic Energy Commission reports that in a dry form, gross fission products are more attractive for industrial application; the specific activity may be as high as 30 curies per gram and they have greater convenience in use and safety in application.

Mixed Fission Products

Mixed fission products are obtained by partial separation of the crude gross fission products into groups of radioisotopes. The reason for the separation is to obtain a group of isotopes with a rather specific type and energy of radiation, and with a rather homogeneous half-life, and high specific activity.

The separation of zirconium-niobium group, which has a half-life of about 65 days and a gamma energy of 0.7 Mev, yields a final product which could be processed to provide a relatively safe and stable solid radiation source. The specific activity could be in the hundred curies per gram range, or about 10 times that of the gross fission products. According to the U. S. Atomic Energy Commission, the separation process does not appear difficult, and preliminary estimates indicate that ZrNb sources might be as inexpensive as four cents per curie, which price would include the cost of source preparation, packaging and shipment for 1,000 miles and subsequent disposal.

Separated fission products

The isolation of specific radioisotopes from gross or mixed products is the final step which leads to the so-called separated fission products. Typical examples are cesium-137 for gamma sources, and strontium-90 as a beta emitter. Cesium-137 emits a gamma ray of 0.66 Mev, useful for radiation applications where product penetration is required. Strontium-90 is a pure beta emitter of 2.18 Mev energy. Its separation from gamma emitting fission products is useful because of the reduction in the severity and cost of shielding and handling precautions. Both cesium and strontium have long half-lives, 33 and 20 years. Their separation from other fission products results in a radiation source which requires relatively little attention over long periods of time thus lowering operating cost projections.

While beta emitters have not received attention as major sources of radiation, their use in radiation processes which have surface treatments as a goal may be satisfactory. While the emphasis on gamma radiation, due to its penetrating power, negated the use of beta emitters such as strontium-90, their application in a broad field involving surface irradiation appears promising. They are competitive with electrons generated by particle accelerators. For those applications where penetration is wasteful and harmful, beta emitters should find applications. Such beta sources show potential advantages in a number of categories and since they are available in good yield from fission products, their use deserves careful appraisal. Massive safe inexpensive beta emitting sources may be fabricated using existing techniques. Source-bearing glass in thin sheets and wire mesh which has been impregnated with the beta emitters are particularly promising. They should be particularly useful for in-stream processing of liquids, and for surface treatments of many biological materials.

Reactor Activated Radioisotopes

Because the elements can be made radioactive by exposure to neutrons, the fabrication of radiation sources of high specific activity and large curie content requires that they be produced in a nuclear reactor, which inherently has a high neutron flux.

The radioisotope so produced of interest in industrial processing is cobalt-60, which emits gamma rays of 1.1 and 1.3 Mev with a half life of about 33 years. Due to the tremendous reactor capacity available in the United States, substantial amounts of cobalt-60 have become available; the production in the United States has yielded megacurie radiation sources. Radioactive sodium loops used as radiation sources, as previously noted, are also reactor made and must be considered as a permanent installation in the reactor site. Cobalt-60 on the other hand may be considered as a portable radiation source.

A report made by the North American Aviation, Inc., on the comparison between reactors and accelerators as sources of radiation for food processing, was summarized as follows. Gamma radiation from nuclear reactors is suitable for radiation processing in depths from 0 to 12 inches whereas particle accelerators are usually limited in depth of penetration. The optimum range for gamma radiation is approximately six inches. At high processing rates (in the range of hundreds of thousands of pounds per hour) a reactor loop gamma source appears cheaper than the cost of particle accelerators required to perform an equal job. As a by-product from reactors, gamma radiation appears competitive with other methods of irradiation.

The problem of the economics of radiation applications will be considered in Chapter 20.

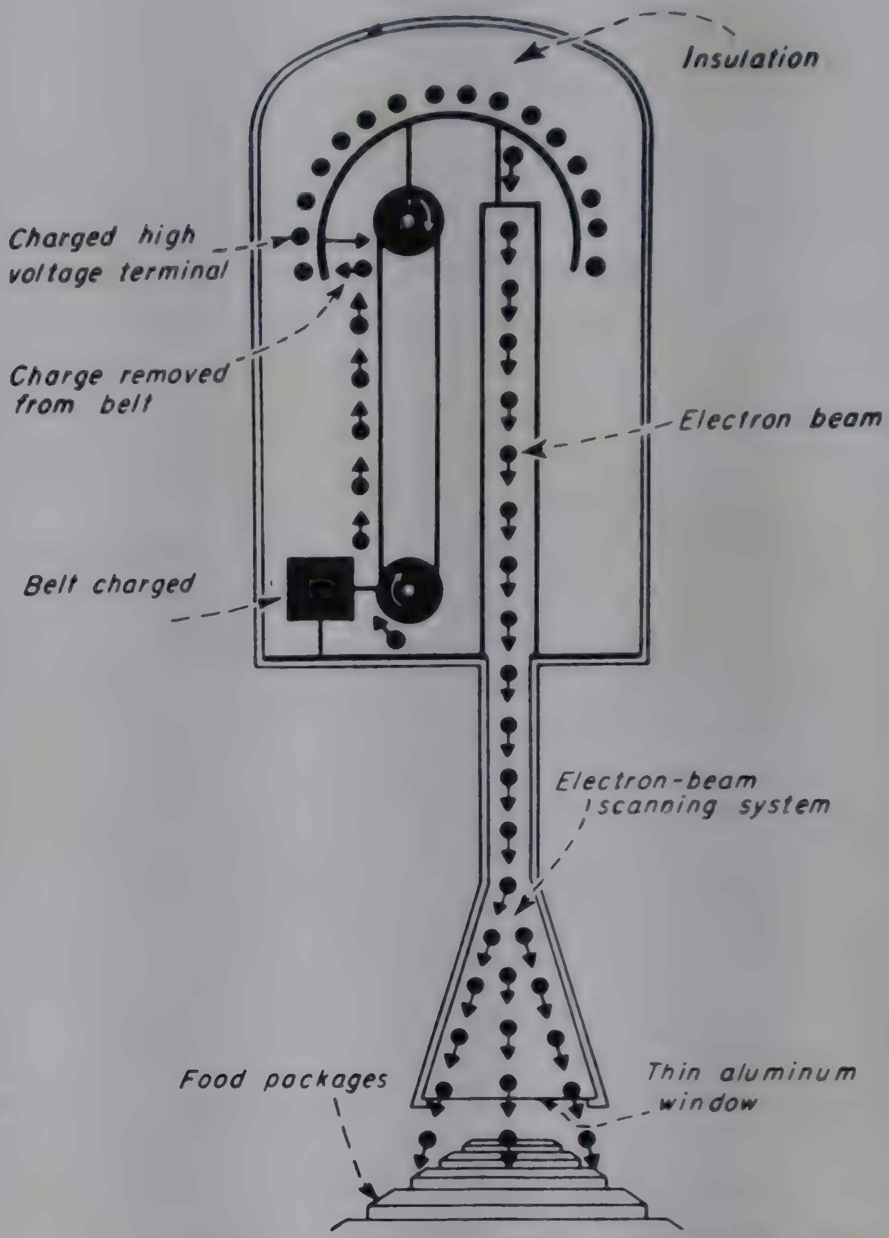
Particle Accelerators

In Chapter 5 the main effect of the interaction of gamma rays with matter was seen to be the production of high energy electrons. As will be subsequently seen, the biological effect of gamma radiation and high energy electrons are, for all intents and purposes, similar. During recent years, machines capable of accelerating particles have made significant progress. The operation of such devices under industrial conditions has proved reliable, and the energy output has been increased (Livingston 1954).

The power of particle accelerators expressed in watts is given by the product of the voltage expressed in volts and the current in amperes:

$$w = v\,i$$

For example, a 1 kw. Van de Graaff generator producing 2 Mev electrons has a beam current of 0.5 milliampere. Or, a cyclotron with a beam current of 1.0 milliampere and a power of 10 kw. emits particles of 10 Mev. In comparison the voltage and beam current in a television tube is


Insulation
Charged high voltage terminal
Charge removed from belt
Electron beam
Belt charged
Electron-beam scanning system
Food packages
Thin aluminum window

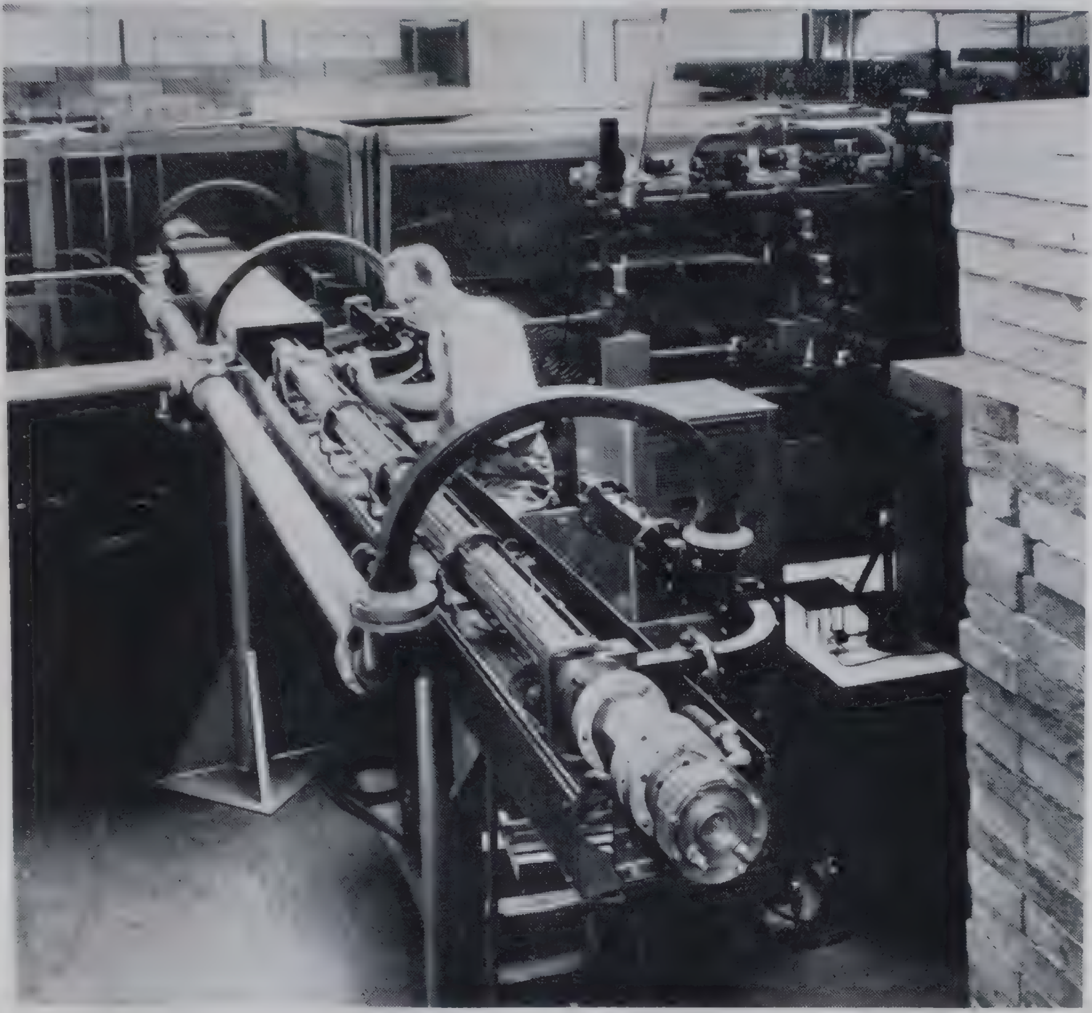
Courtesy of U.S. Army Quartermaster Corps

FIG. 20. SCHEMATIC DIAGRAM OF VAN DE GRAAFF ELECTION ACCELERATOR

of the order of 5,000 volts and 0.01 milliamp, equivalent to a beam power rating of 0.05 watt and 5 Kev electrons.

The large scale artificial production of radiation awaited the development of equipment capable of producing voltages sufficiently high to deliver energies to electrons or protons and carry them forward at accelerated speeds. Cockcroft and Walton in 1930, using vacuum tubes in a voltage multiplying circuit, developed an apparatus which attained an output of 300,000 volts. In operation it delivered a beam of protons which, on impact with a lithium or beryllium target, gave an appreciable yield of alpha particles. This and similar machines were useful in studies of nuclear disintegration but were extremely bulky, expensive, and the samples irradiated were limited to extremely small sizes.

In 1931, Van de Graaff developed a high voltage electrostatic generator capable of a steady output. Essentially the Van de Graaff unit consisted of a large metal dome supported by an insulated column. Inside this column was an endless belt of non-conducting material, which transported an electric charge from the electron source to the dome. The dome then acquired a high voltage with respect to ground. An excess of 10,000,000 volts has been obtained and utilized in accelerating electrons through a long cylindrical vacuum tube and out through a thin aluminum window. The source of electrons was a heated filament. The high voltage was applied between the electron source and the window. By employing an oscillating magnetic field at the base of the vacuum tube, it is possible to move the electron beam back and forth over the area being bombarded. This feature would allow the processer or industrial user to treat material uniformly, under the beam, on a conveyor in a continuous operation. Such a device developed by the High Voltage Engineering Corporation is shown in Fig. 21. Van de Graaff machines are currently available commercially



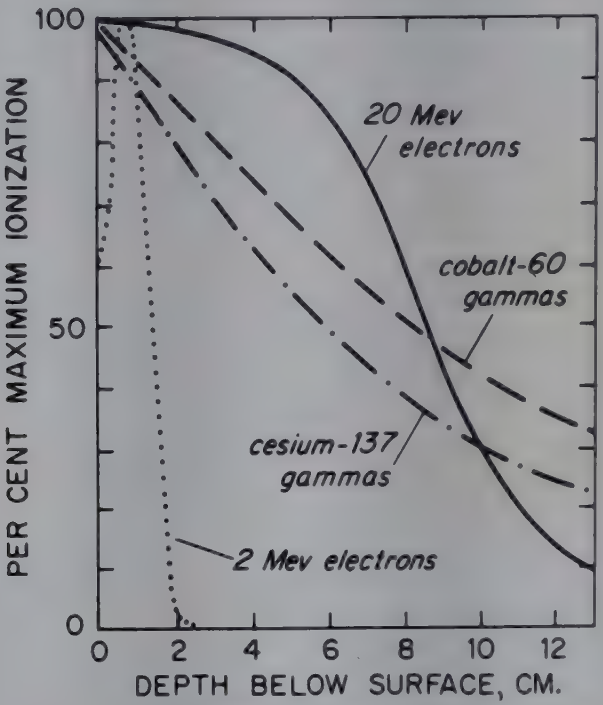
Courtesy of High Voltage Engineering Corp.

FIG. 21. LINEAR ACCELERATOR, 50 MEV ELECTRONS

with electron energies ranging from 1.0 Mev to greater than 3 Mev with beam power ratings from 250 W to 5,000 W.

Linear Accelerators

Another source of high energy electrons, the linear accelerator, was proposed. This consisted of a horizontal evacuated cylinder with an electron source at one end and a target at the other, either within the cylinder or just outside a thin aluminum window through which the high velocity electrons emerged. Linear accelerators do not use the ultra-high voltages for accelerating particles, but instead a traveling wave at high frequency power generated and driven down the evacuated cylinder. Actually bunches of electrons are fired into the cylinder, are trapped by the electromagnetic field associated with the wave, then accelerated to the speed of the wave itself.

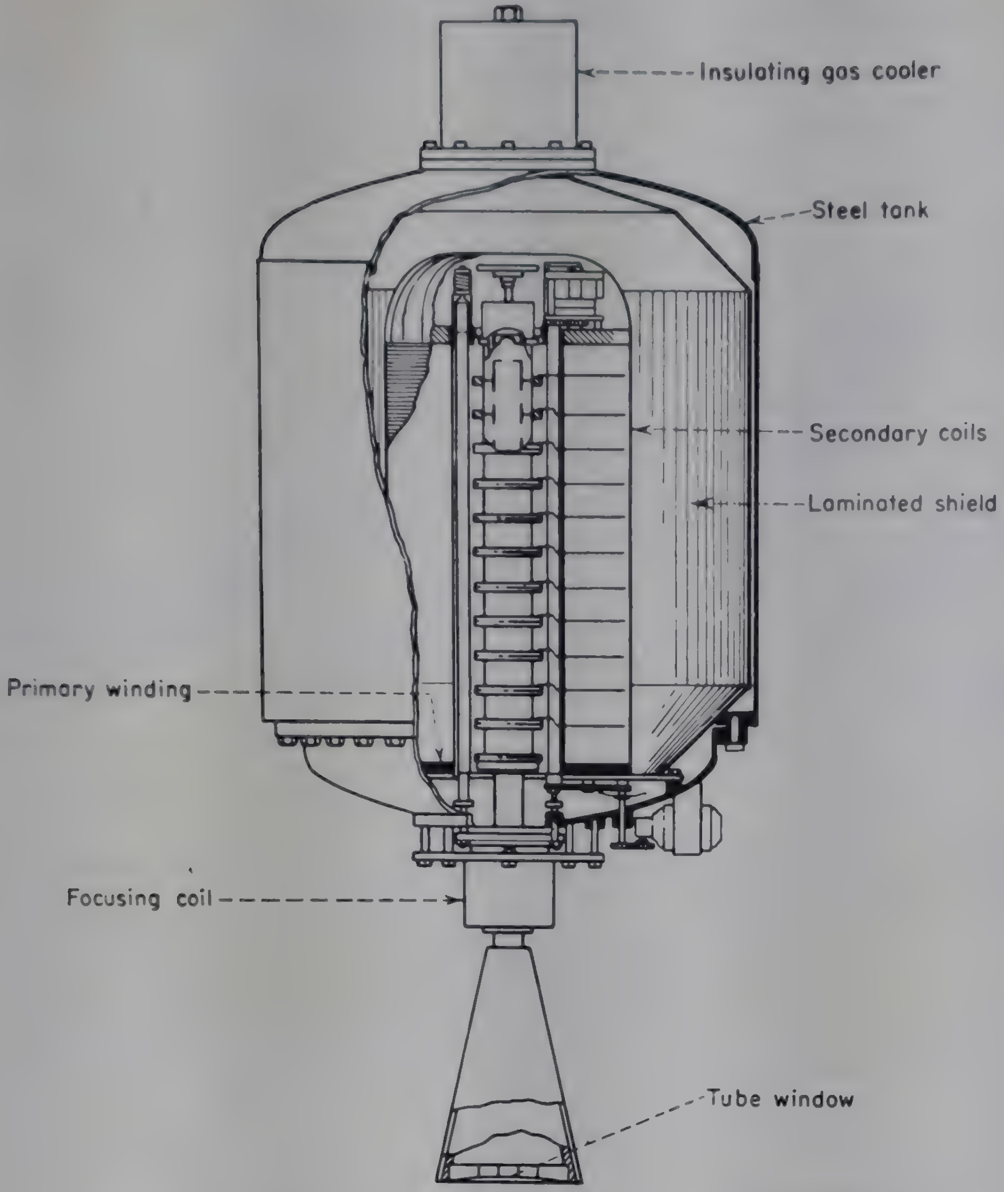


Courtesy of U.S. Army Quartermaster Corps. From Gale (1958)

Fig. 22. Dose-Depth Relation for Electrons and Gamma Ray Sources

The output energy of the first machine was 1.2 Mev. Technical improvements have recently raised this energy. Fig. 21 shows a 50 Mev unit developed by the High Voltage Engineering Corporation. The penetration of electrons into water or like material is shown in Fig. 22; the penetration is approximately 0.5 cm. per Mev of beam energy.

While the electrostatic (Van de Graaff) accelerator delivers a constant current of electrons of uniform energy, the linear accelerator delivers a series of current pulses, each of microsecond duration, at repetition rates


Insulating gas cooler
Steel tank
Secondary coils
Laminated shield
Primary winding
Focusing coil
Tube window

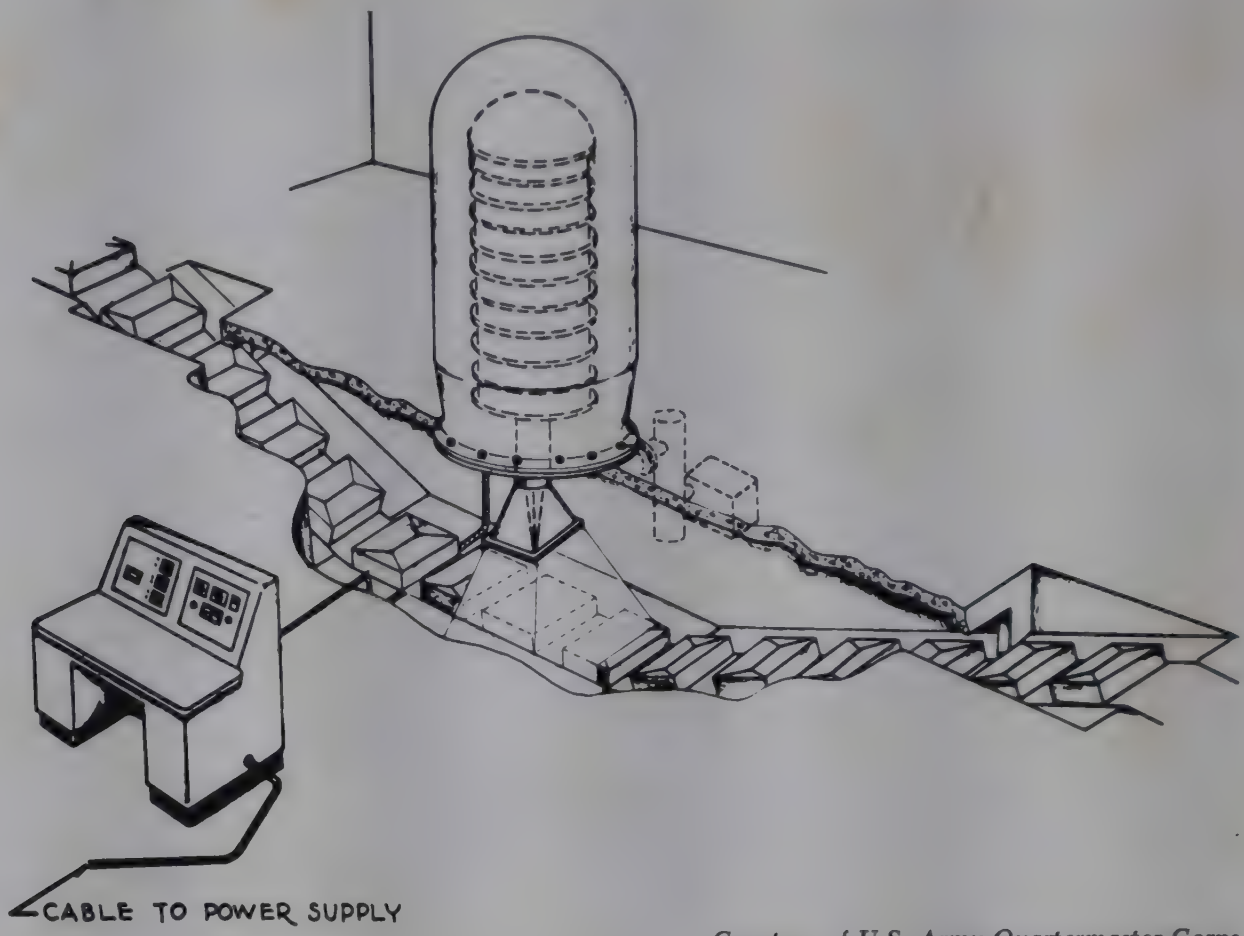
Courtesy of U.S. Army Quartermaster Corps

FIG. 23. SCHEMATIC DIAGRAM OF RESONANT TRANSFORMER

in excess of 500 pulses per second. The beam power may be varied by changing the pulse repetition rates, while keeping the electron energy fixed. The scanning devices and techniques also apply to the linear accelerator. Further, the interchangeability of components has been introduced which give a variable electron energy output.

Resonant Transformer

High voltage X-ray technology developments during the past few years have made available devices for the production of high energy electron beams. In this case a secondary or high voltage circuit on the X-ray transformer is tuned to resonance, thereby eliminating the need for an iron core. This yields advantages in weight and size. Such devices are currently available with energies ranging from 1 to 3.5 Mev and beam power



Courtesy of U.S. Army Quartermaster Corps

FIG. 24. SCHEMATIC DIAGRAM OF CAPACITRON

ratings upwards to 26,000 W. Such a device is shown schematically in Fig. 23.

Capacitron

This device produces high electron accelerating voltages and high beam power during micro-second time periods. A number of capacitors is charged in parallel through resistors from a rectifier unit. The capacitors are discharged in series by means of gaps which spark over at predetermined voltages. While such devices are capable of generating 5 Mev electrons they have not yet been employed to the same degree as some of the other equipment. This machine is shown in Fig. 24.

Dynamitron

This device converts low voltage a-c power to high voltage d-c power by means of a cascaded rectifier system driven in parallel from a radio frequency oscillator. The rectifier array is located between, and spaced apart from, a pair of large curved electrodes which form the tuning capacitance of the oscillator. Strong alternating fields between the electrodes induce currents through the series connected rectifiers, building up a large constant potential at the end of the array. The Dynamitron is capable of delivering large electron beam currents at potentials upwards of six Mev.

X-ray Generators

X-rays are produced by bombarding heavy metal targets with a beam of high velocity electrons. Recent developments have been concentrated on very high current, low voltage X-ray machines, potentially useful for irradiation. Because of the low energy conversion efficiency, however, X-rays would appear useful for applications not requiring large doses or deep penetration in substrates. There are, of course, well-established uses for X-ray equipment for medical diagnostic radiography and radiation therapy as well as industrial radiography. However even here X-ray machines are being challenged by other radiation sources, particularly portable cobalt-60 gamma sources.

FUNCTIONAL RADIATION SOURCES

Of the particular types of ionizing radiations available, gamma and beta radiation from radioactive elements, and machine generated electron beams are the most applicable for processes of industrial import. The use of machine-produced ionizing radiation offers certain distinct advantages over those available from fission products.

Gamma rays for widespread industrial radiation applications as discussed above are available from radioisotopes either from fission products or reactor activated nuclides. Beta rays are also available from fission products and reactor activated nuclides. Electrons are available from a number of functional accelerating devices.

The disadvantages in the use of fission products are that they require extensive shielding and the dose applied may be low thereby necessitating long exposure times.

On the other hand, electrons have low penetrating power when compared to gamma rays. Depending on the application, low penetrating ability may be either an advantage or a disadvantage. For surface sterilization or treatment, it may be an advantage, putting the energy where it is desired. Machine produced beams are unidirectional; further they can be turned off!

There is no reason why shielding cannot be a material to be treated, e.g., water, a major component of biological substrates; it should be possible with knowledge and imagination to design an aqueous system which in the process of absorbing the dangerous radiation of a reactor, or other radiation source, undergoes chemical changes which form useful materials which can be separated, packaged, and marketed. In analogy the breeder reactor is actually a dual purpose device.

CHARACTERISTICS OF RADIATION

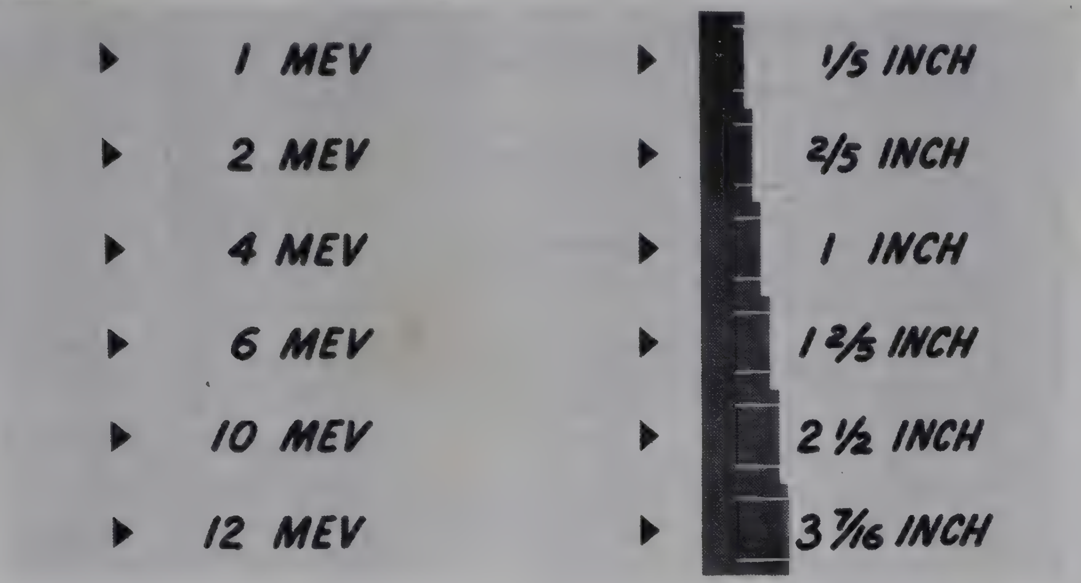
Electromagnetic Radiation

In review, X-rays are produced by the bombardment of a heavy metal with a beam of high velocity electrons. Much more penetrating than ultra-violet or ordinary light, X-rays, however, cannot readily be focused, bent, or scanned. The source-to-product radiation geometry is therefore inflexible and results in a relatively low efficiency (20–40 per cent) of use of the radiation produced. The absorbed dose is always a fraction of the total radiation power produced by a source. X-rays with less than two Mev energy do not induce radioactivity in an irradiated substrate. Above two Mev some significant activity may occur by the photo-neutron reaction previously discussed in Chapter 5. Because of the poor conversion efficiency of electron energy to X-ray energy, under ordinary circumstances, the applications appear limited. At two Mev approximately five per cent conversion occurs (Pomeranz, Siu, and Evans 1957). The efficiency can be increased to about 30 per cent at energies ranging from 15 Mev, to 45 per cent at 25 Mev. The cost of X-rays absorbed per unit of substrate is therefore high at the lower end of the electron energy spectrum. At the higher end, the penetrating range of electrons themselves is sufficiently great to satisfy many of the industrial applications for which X-rays might themselves have been considered.

Gamma rays are similar to X-rays, but of shorter wave length. Gamma rays are photons of energy emitted from radioactive nuclei, distinct from X-rays emitted by the bombardment of heavy metal targets. The gamma ray energies available from cesium-137 and cobalt-60 range from 0.7 to 1.3 Mev, and greater product penetration is available, conveniently, with gamma rays than with X-rays. Like X-rays, gamma rays also cannot be focused, bent, or scanned. Here again the source-to-product irradiation geometry is somewhat inflexible and results in radiation use efficiency of the order of 20 to 40 per cent.

The problem of increasing the useful geometry of a source is difficult, involving severe problems of materials handling, shielding, maintenance, and replacement. The efficiency figures listed above are those currently available, yet it appears very likely that imaginative concepts of source geometry design will improve them.

Like X-rays, gamma rays in excess of 2.3 Mev energies and particularly above 8 Mev can induce radioactivity in an irradiated substrate by interacting with the deuterium isotope of any hydrogen present causing it to emit a neutron (photonuclear reaction) which can then induce radioactivity in any neutron-activatable element present in the product. An example is seen in the activation of potassium, a common constitutent of living matter. Gamma sources emit their radiation in all directions, and therefore require extensive shielding or imaginative use of the radiation field. This poses difficulties in maintenance of radiation sources, and such sources must usually be physically closed from the scene of operations. This may be overcome with sources which may be easily moved (for instance, lowered into a water well, a generally employed practice). With stationary sources such as would be found in loops through which coolants from reactors pass, equally elaborate procedures are required. Large gamma sources usually require cooling because of self-absorption of the gamma or beta radiation present and must be constantly monitored to prevent or at least detect leakages of radioactive materials. Gamma sources must also be replaced at intervals, depending upon their half-lives, to maintain a uniform source strength.



Courtesy of U.S. Army Quartermaster Corps

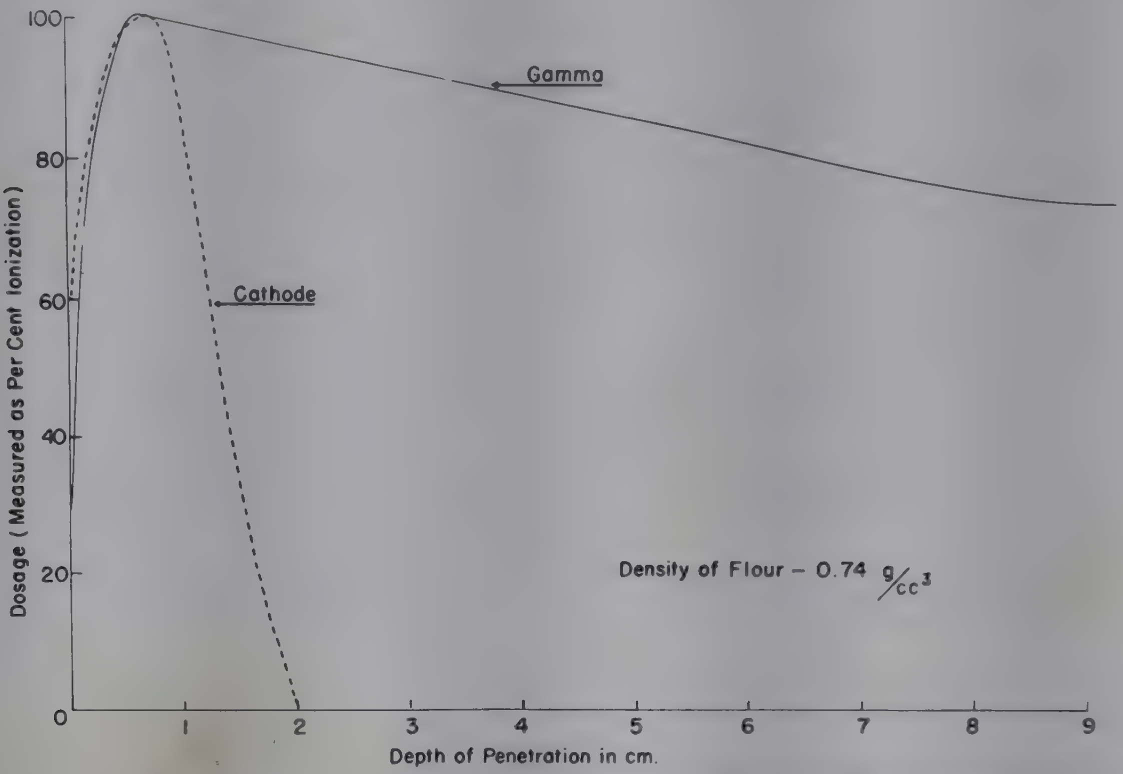
FIG. 25. PENETRATING CHARACTERISTICS OF ELECTRON BEAMS

It has been found useful, therefore, to have multiple units in an array, to facilitate replenishment. The penetration of gamma radiation in comparison to the penetration of high energy electrons is shown in Figs. 22, 25, and 26. The depth of penetration and the uniformity of treatment are therefore important considerations in the selection of suitable radiation sources.

Also one should investigate the possibility of establishing production or irradiation schedules which are compatible with the isotope half life. There are many processes for which the product demand exhibits regular, predictable variations with time.

Particle Radiation

Alpha particles, neutrons, protons, deuterons, and positive ions are all classified as heavy particles. Some lack penetration ability, for example alpha particles, while others such as neutrons are too penetrating, hence not easily absorbed. Several of these induce radioactivity in a treated substrate. All are relatively expensive to produce. While neutrons are useful, they have severe limitations due to neutron induced radioactivity in treated material.



Courtesy of U.S. Army Quartermaster Corps

FIG. 26. DOSAGE DISTRIBUTION BY 3 MEV CATHODE (ELECTRON) AND GAMMA RAYS IN FLOUR

Beta particles are high energy electrons emitted by many radioactive elements. A one Mev beta particle penetrates approximately 0.5 cm. in water. This enforces a limitation in comparison to the penetrating ability of gamma rays. However, when applications are made where penetrating power is not a significant problem, beta applications are useful. However, as is the case with gamma sources, beta sources must be safely contained and monitored for leakage. Shielding requirements are less than for gamma sources but must also be effective in all directions. Ingenuity in source design may be an important factor in the uses which beta rays find as noted earlier.

Electrons are the same as beta particles, the term beta being restricted to those emissions of electrons from a radioactive element. Electron beams are usually associated with high energy accelerating equipment as previously discussed. Accelerators are able to yield electron beams upwards of 25 Mev, which find varied industrial processing applications. Penetration ability at these higher energies is substantial, but above 10 Mev there is the problem of inducing radioactivity in the treated material by direct activation of elements with high energy electrons. Even below 10 Mev, some trace activity can be induced by the conversion of energy through bremsstrahlung to neutron activation, discussed in Chapter 5.

Because electrons can be focused, bent, and scanned, highly intense beams can be directed uniformly to substrates. Beam-splitting techniques have been applied successfully to a substrate by bombarding uniformly from two or more sides simultaneously to provide uniform distributions of energy. As indicated earlier, shielding of accelerators is less a problem than that for radioactive isotopes due to the usually well-defined emerging beam. However, shielding from secondary radiations, such as bremsstrahlung, must be considered.

General Source Requirements

To successfully control a radiation process, it is necessary to have information not only on the distribution of energy by the source, but also information on the energy absorbed by the substrate being treated.

Source strength gives only a measure of the number of emitted radiations per unit time. The quality of the radiation together with the number of radiations gives a measure of the energy flux. This may be shown in comparing the curie of cobalt-60 with a curie of tritium. Cobalt-60 yields more energy flux than the tritium. However the beta particles emitted by tritium are much more readily absorbed than the cobalt-60 radiation, and an exposure of a substrate to the two sources for an equal length of time in equivalent geometries may find more absorption of energy from the tritium source than from the cobalt source. Another illustration is found in the irradiation of a gram of butter and a gram of lead, both exposed to the same gamma flux from cobalt-60. The lead would absorb a greater energy total since the radiation interaction is almost exclusively with the electron surrounding the nuclei. Lead has substantially more electrons per cubic centimeter than does butter.

The dosage distribution versus penetration in flour by cathode (electron) and gamma radiation is shown in Fig. 26.

Studies of the distribution of radiation energy absorbed in a container are shown in Figs. 27A and 27B (Herschman 1956). In Fig. 27A, a mount is shown onto which are placed a number of cobalt glass dosimeter strips discussed in the next chapter. It is possible to arrange these dosimeters spatially so that when placed in position in the container shown in Fig. 27B, then irradiated, a three dimensional map is obtained indicating the distribution of energy absorbed throughout the sealed container while exposed in the radiation chamber. Nature places an absolute maximum on the uniformity of dose distribution which can be achieved, and process economics dictates further compromise. In any particular radiation field, as mapped, the presence of irradiated material in an array itself alters the radiation field and can yield non-uniformity of dose absorbed. Experimental data indicate that variations of at least 10 per cent are encountered. Such information can be obtained by judiciously placing cobalt glass dosimeter strips in agar gels of desired size and shape for irradiation. Another method employs methylene blue dye in agar gels. Color changes in the dye map the distribution of radiation in three dimensional patterns.

Extensive studies indicate that variations in dose distribution in many types of containers and with many types of sources can be controlled to within about 25 per cent providing the container size is selected for the particular radiation source. If successful processing is to be achieved, no less than 100 per cent of the desired dose must be administered to each container. Inasmuch as the radiation dose is directly correlated to the changes in the substrate, variations must be maintained at a minimum. The range of 100 to 125 per cent of dose required appears tolerable in most substrates, and appears to be compatible with radiation source performance and the economics of the process. The above considerations apply particularly to food sterilization.

TABLE 12

MAJOR RADIATION SOURCES AVAILABLE

Source	Radiation Obtained
Nuclear Reactor	
Reactor Core	Neutrons (fast and Thermal), gamma and fission fragments
Fission Products	
Spent Fuel Elements	Gamma
Gross Fission Products	Gamma and beta
Mixed Fission Products (such as ZrNb)	Gamma and beta
Separated Radioisotopes (such as cesium)	Gamma and/or beta
Activated Radionuclides (such as cobalt-60)	Gamma and/or beta
Activated Radionuclides (such as sodium in reactor loops)	Gamma
Particle Accelerators (such as Van de Graaff, linear accelerator, resonant transformer, Capacitron, Dynamitron, Cockroft-Walton)	Electrons (beta, protons, deuterons, alpha, and positive ions)
Targets of accelerators (including x-ray tubes)	X-rays, neutrons
Electric Discharges (such as spark, glow, corona, high-frequency)	Electrons
Ultra-violet light	Low-energy photons

Both high energy electromagnetic radiations (e.g., gamma rays) and high energy particles (e.g., high energy electrons) are capable of promoting chemical, physical, and biological reactions of interest. To be of practical value in radiation technology, the radiation chosen should:

1.—penetrate the substrate to be treated with reasonable uniformity and be of such a type that most of the imparted energy is absorbed in the irradiated sustrate.

2.—induce no objectionable radioactivity in the irradiated substrate or at most, short-lived species.

3.—be reliably controlled, safe in operation, and economical in shielding.

4.—be obtainable from radiation sources which provide an adequate output at reasonable costs.

The major radiation sources available are summarized in Table 12.

BIBLIOGRAPHY

Coryell, C. D., and Sugarman, N. 1951. Radiochemical Studies: The Fission Products. McGraw-Hill Co., New York.

Friedlander, G., and Kennedy, J. W. 1955. Nuclear and Radiochemistry. Second Ed. John Wiley and Sons, Inc., New York.

Gale, A. J. 1958. Ionizing radiation. Amer. Soc. Plastics Engr. 14th Annual Conference *4*, 55–63.

Glasstone, S. 1958. Sourcebook on Atomic Energy. Second Ed. D. Van Nostrand Co., Inc., Princeton, New Jersey.

Hahn, O., and Strassmann, F. 1939. On the detection and behavior of the alkaline earth metals formed by the neutron irradiation of uranium. Naturwissenschaften 27, 11 (in German).

Harteck, P., and Dondes, S. 1956. Producing chemicals with reactor radiations. Nucleonics *15*, No. 8, 94–96.

Herschman, A. 1956. Induced radioactivity considerations in foods. QM. Food and Container Institute Research Report. Proj. 7-84-01-002. File No. 0203AD. Chicago Ill.

High Voltage Engineering Corporation. 1956. Van de Graaff Particle Accelerator, Bull., E. Cambridge, Mass.

Livingston, M. S. 1954. High Energy Accelerators. Interscience Publishers, New York.

Meitner, L., and Frisch, O. R. 1939. Disintegration of uranium by neutrons: a new type of nuclear reaction. Nature *143*, 239.

North American Aviation, Inc. 1956. Logistics and economic feasibility study on radiation sterilization of foods. Final Report U. S. Army QM. Corps. Contract No. DA 19-129-QM 491.

Pomeranz, R., Siu, R. G. H., and Evans, B. S. 1957. Sources of ionizing radiation, Chap. 29. Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

Proceedings of the First International Conference, Peaceful Uses of Atomic Energy. 1956. United Nations, Geneva. 16 volumes.

Proceedings of the Second International Conference, Peaceful Uses of Atomic Energy. 1958. United Nations, Geneva. 33 volumes.

United States Army QM. Corps. 1957. Radiation Preservation of Food. U.S. Govt. Printing Office, Washington, D.C.

VAN DE GRAAFF, R. J. 1931. A 1,500,000 volt electrostatic generator. Phys. Rev. *38*, 1919.

WEINBERG, A. M., and WIGNER, E. 1958. The Physical Theory of Neutron Chain Reactors. University of Chicago Press, Chicago, Illinois.

WEST, J. M., and ROY, G. M. 1959 The future of boiling-water reactors. Nucleonics *17*, No. 1, 42–49.

WALLENSTEIN, M. B., ROSENSTOCK, H. M., and JOHNSTON, W. H. 1959. Rept. on Fundamental Considerations in Radiation Technology. JLI 650-3-3, AEC Contract AT(11-1)-650.

CHAPTER 7

Measurement of Radiation Dose

Radiation dosimetry involves the measurement of the energy actually absorbed in a substrate exposed to ionizing radiation. The radiation may be either electromagnetic or corpuscular in nature. As has been seen in Chapter 5, the passage of electromagnetic radiation through a substrate causes ionization by virtue of the secondary electrons released. Particle radiations on the other hand, ionize directly because they are charged, or indirectly through the charged particle resulting from a collision with a substrate. Alpha, beta particles, and protons yield primary ionization. Gamma rays and neutrons produce ionizing radiation by their interaction with the substrate in passage through it.

Measurement of radiation dose involves the intensity of the source (characteristic solely of the source), the cumulative effect on the substrate, and the rate at which the effect is brought about. The source is characterized by the nature and energy distribution of the radiations, and the rate at which the radiation is being emitted (1 curie = 3.7×10^{10} disintegrations per second).

Units of Radiation Dose

The original unit of measurement of radiation was the roentgen, r, and was defined in terms of ionization events. A roentgen (r) of radiation has been defined as:

1. The quantity of radiation which produces one esu (electrostatic unit) of positive or negative electricity per cubic centimeter of air at standard temperature and pressure, or,

2. The quantity of radiation that will produce 2.083×10^9 ion pairs per cubic centimeter of dry air, or,

3. The radiation received in one hour from a 1 gram source of radiation at a distance of one meter.

A single charged ion carries 4.8×10^{-10} electrostatic units and one r forms, in one cubic centimeter of air, 2.08×10^9 ions of either sign, or 2.08×10^9 ion pairs. At standard conditions one cubic centimeter of air weighs 0.00129 gram so that one gram of air absorbing one r would result in the formation of 2.08×10^9 divided by 0.00129 or 1.61×10^{12} ion pairs. The energy required to produce one ion pair in air is known to be about 32.5 electron volts. The energy required to form 1.6×10^{12} ion pairs is 5.23×10^{13} electron volts. Since one electron volt is equal to

1.60×10^{-12} ergs, the energy gained by absorption of one r in one gm. of air is about 89 ergs, for gamma and X-rays. This unit was designated for these radiations. Since the biological effects due to ionization from alpha, beta, and proton particles are similar in tissues, the term r has been extended to these radiations in addition to photons of electromagnetic radiations. The determination of a dose in roentgens requires a measurement of an ionization current under saturation conditions based upon the theory of the cavity ionization chamber.

The method does not give a direct measurement of the absorbed dose. It actually yields a measurement of the amount of the charge produced by a given exposure to a particular energy of radiation absorbed in the ion chamber. The results of many experiments have led to the conclusion by many workers that for dry air, the roentgen implies an energy absorption of 89 ± 1.3 ergs per gram of dry air. For gases other than air, the energy absorption per r depends upon the number of electron volts expended in forming an ion pair and the density of the gas.

In the case of liquids, ion pairs may be produced, but they recombine very easily and quickly. Direct experimental measurement of the amount of charge produced in a liquid is virtually impossible under such circumstances. The reliability of the roentgen is also questionable when secondary corpuscular radiation is not in equilibrium with the primary ionizing source. At an air-solid or an air-liquid interface where the ionizing radiation must reach a finite depth before reaching equilibrium with secondary ionization, the use of the roentgen is again inadequate. Because of such shortcomings, a number of units have been proposed for the evaluation of the radiation absorbed by a substrate. These include the gram roentgen, the roentgen equivalent physical, the roentgen equivalent man, and the rad.

The gram roentgen is the amount of radiation absorbed per gram of dry air. This is a quantity equal to 89 ± 1.3 ergs per gram. Because the radiation absorption characteristics of moist tissue are different from that of air, the energy absorbed by the tissue would be greater.

The roentgen equivalent physical (rep) was developed to describe the dose rate for primary ionizing radiations, e.g., high energy electrons. The rep is defined as that dose of ionizing radiation which produces an energy absorption of 84 ergs per cubic centimeter of tissue. This value was later adjusted to 93 ergs per cubic centimeter. The roentgen equivalent mammal (rem) is defined as that dose absorbed by a mammal when exposed to ionizing radiation biologically equivalent to the dose of one r of gamma radiation. The dose in rem is equal to the dose in rep multiplied by the relative biological effectiveness (RBE). The rem is of little value unless the RBE is specified and applied.

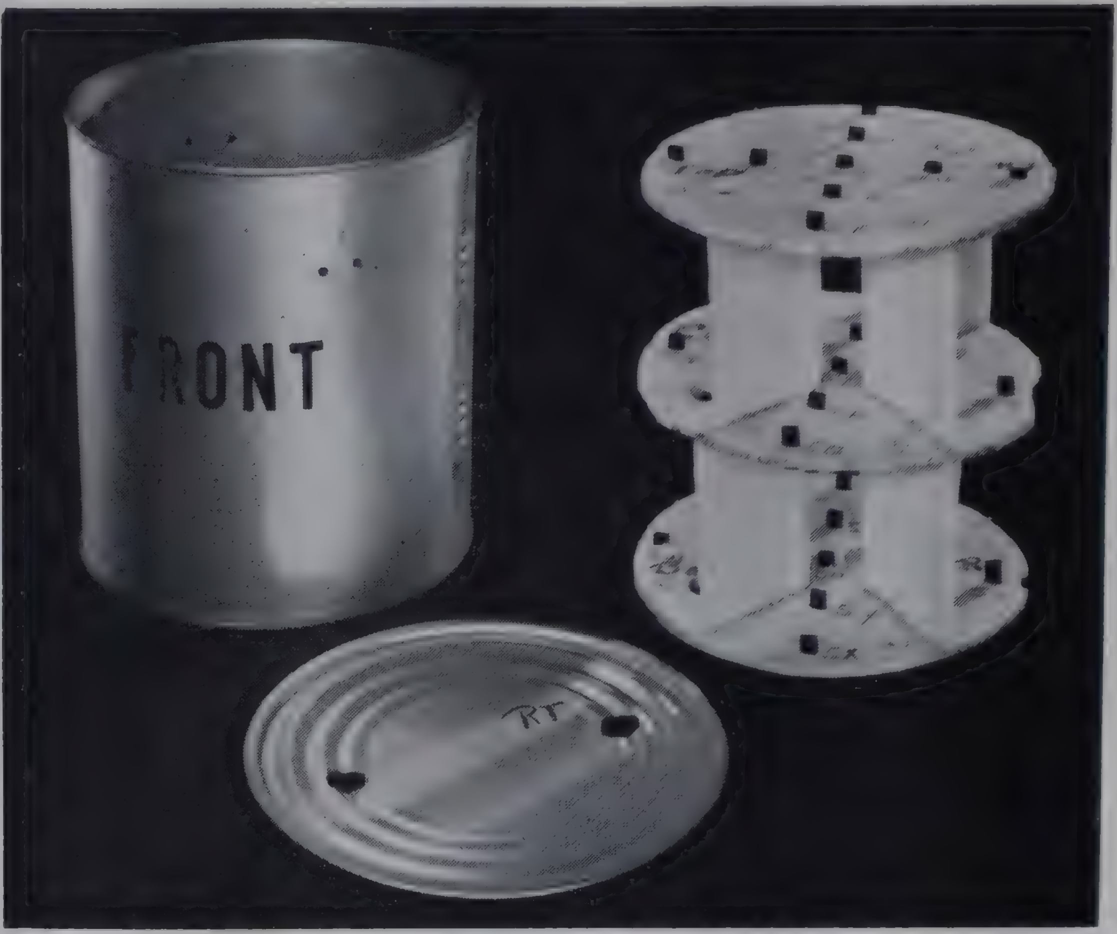
The rad is the unit currently in widespread use which defines the absorption of energy by a substrate directly. The rad is defined as the absorption of 100 ergs per gram of material. This yields a direct means of measurement which has been found most useful in radiation technology. The rad is approximately 1.1 times larger than the rep found in the literature. The current trend in radiation technology favors the use of the rad unit.

Dosimeters

To be functional, a dosimeter should be independent of the dose rate over a wide range and be relatively insensitive to small temperature and pressure changes. There should be no dependence upon the concentration of materials in the substrate, as they may change during irradiation. Furthermore, the quality of the radiation applied should not be a determining factor in the response of the dosimeter. It would be desirable to have simple procedures involving no special purification of needed reagents, and solutions used in dosimetry should be usable in equilibrium with natural surroundings. Chemical dosimeters most often fail in the requirement of independence of the quality of radiation supplied. Nonchemical dosimeters often fail in the quality of dose rate, temperature and pressure response, as well as the quality of radiation. Dosimeters have therefore evolved having unique features of special application. Unfortunately functional, simple, durable dosimeters capable of evaluating radiations varying in intensity a millionfold, are not available. However useful dosimeters are available for various dose ranges and will be described below.

The establishment of primary standards for the measurement of radiation dose is complex. However, once primary standards are established, secondary references can be devised which usually are more convenient than primary standards.

Dosimeters of three types are actually required; primary standards, operating standards, and production control devices. An example of a primary standard dosimeter is the calorimeter developed at the National Bureau of Standards. Usually primary standard dosimeters do not lend themselves to routine usage, and operating dosimeter systems have been developed which have been standardized against primary dosimeters. Modified ionization chambers can be used to monitor a radiation source continuously. The operating standard dosimeters found useful in routine radiation technology are cobalt glass, ferrous sulfate (Fricke 1928), and ceric sulfate dosimeters. The ceric sulfate dosimeters are functional for doses between 200,000 and 5,000,000 rads; the cobalt glass dosimeters are



Courtesy of U.S. Army Quartermaster Corps

FIG. 27A. DOSIMETRY WITH COBALT GLASS STRIPS

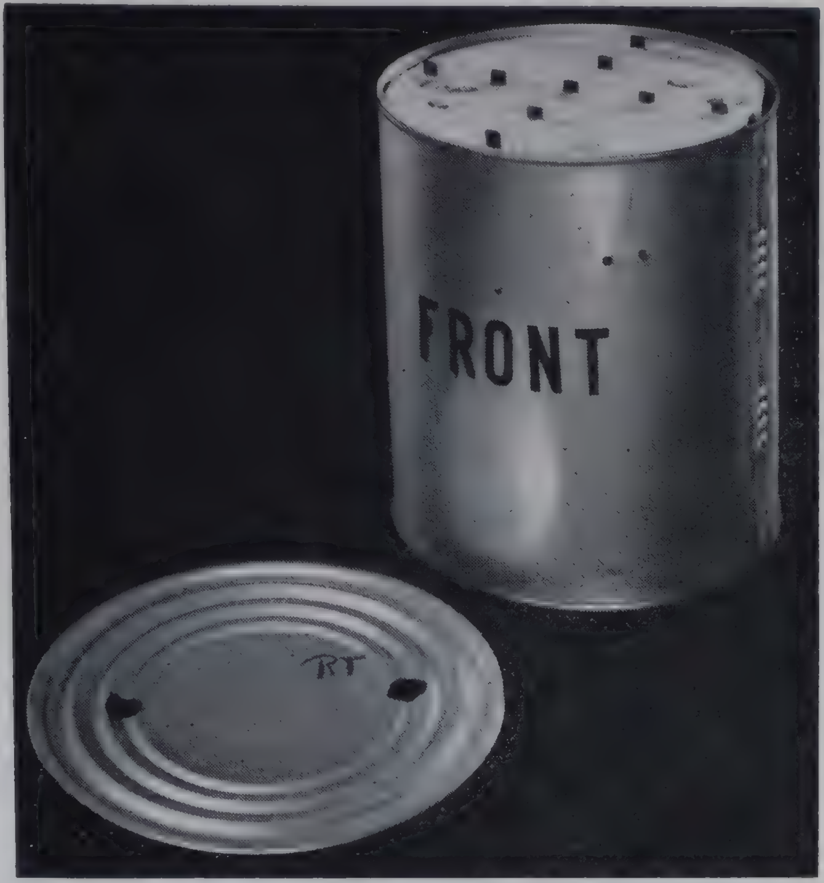
Plastic holder used to study dose distribution

operative between 200,000 and 10,000,000 rads; and the ferrous sulfate dosimeters are functional below 50,000 rads.

Ferrous Sulfate Dosimeter.—One of the most widely used standards is the Fricke dosimeter, which consists of an acid solution of a ferrous salt containing some chloride to inhibit side reactions and impurity effects (Fricke and Morse 1928).

The formation of ferric ions is the key effect which permits the evaluation of the radiation dose; the concentration of the ferric ion is determined spectrophotometrically by its absorption band at a wave length of 305 millimicrons. The concentration dependence of the light absorption is exponential (Beer's Law), so that the logarithm of the light attenuation is proportional to the concentration. On the basis of careful experimentation, it has been found that there are 15.4 ± 0.1 ferric ions formed for every 100 electron volts of absorbed energy, so that the determination of the ferric ion concentration gives a direct measure of the energy absorbed. The yield of ions per 100 electron volts is called the G value, and in this case G equals 15.45. A detailed method for the use of the Fricke dosimeter is given in reference. The mechanism is discussed in Chapter 8.

Ceric Sulfate Dosimetry.—The ceric-cerous dosimeter system is influenced by the same general and specific factors which influence the Fricke dosimeter. However, the ceric system is not influenced by the oxygen content, nor by dose rate effects. The ceric ion concentration may be determined either by titration with ferrous sulfate or by direct measurement spectrophotometrically of the absorption of ceric ion at 310 millimicrons. The latter method is generally used. The procedure for using ceric sulfate in dosimetry is presented in reference (Harlan and Hart 1959).

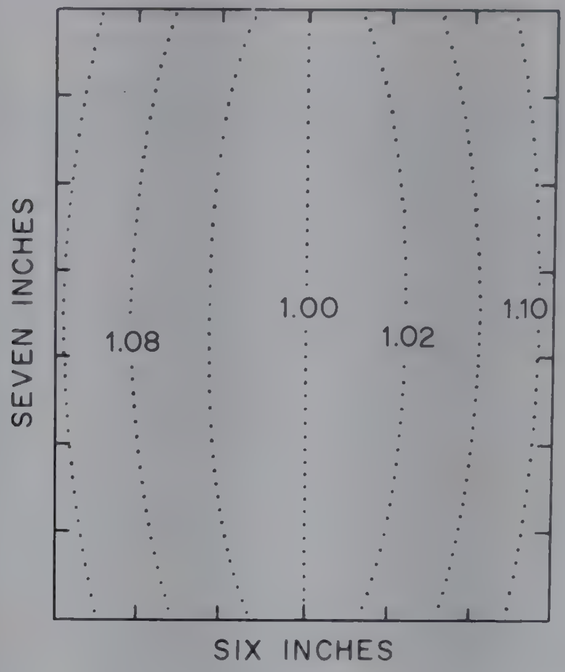


Courtesy of U.S. Army Quartermaster Corps

FIG. 27B. DOSIMETRY WITH COBALT GLASS STRIPS

Dosimeters sealed inside container and sent through radiation source

Cobalt Dosimeter Glass.—High level dosimetry of gamma radiation has been found functional by the use of cobalt glass dosimeters (Figs. 27). A silicate glass containing 0.1 per cent Co_3O_4 is manufactured by the Bausch and Lomb Optical Company, Rochester, New York, and is available from the manufacturer. The chief advantages in using the cobalt glass system over other means of dosimetry are the small size, ruggedness, and ease of mounting for mapping dose distributions, and the possible reuse by heating to eradicate the color developed during the radiation. Because glass tends to fade during storage, the system must be calibrated before use and optical values compared with some absolute primary system of dosimetry such as the ion chamber, the calorimeter, or even a ceric sulfate system. The usable dose range is usually considered above 200,000 rads and below 10,000,000 rads. Glass dosimeters are capable of high precision, in the realm of two per cent. Heating such glass for six hours at 450°C. in a muffle furnace permits re-use of the glass dosimeter.



Courtesy of Rest

FIG. 28. ISODOSE CURVES FOR 6 BY 7-INCH CYLINDER IN GAMMA FIELD

The use of sandwiches of glass five layers thick is useful in establishing electronic equilibrium in the system thereby making it independent of the surrounding material. The assumption is made that the measurements are made with essentially one Mev photons which cause secondary electrons to be emitted which will have a range of under 1.5 mm. in the glass (Kreidl and Blair 1956).

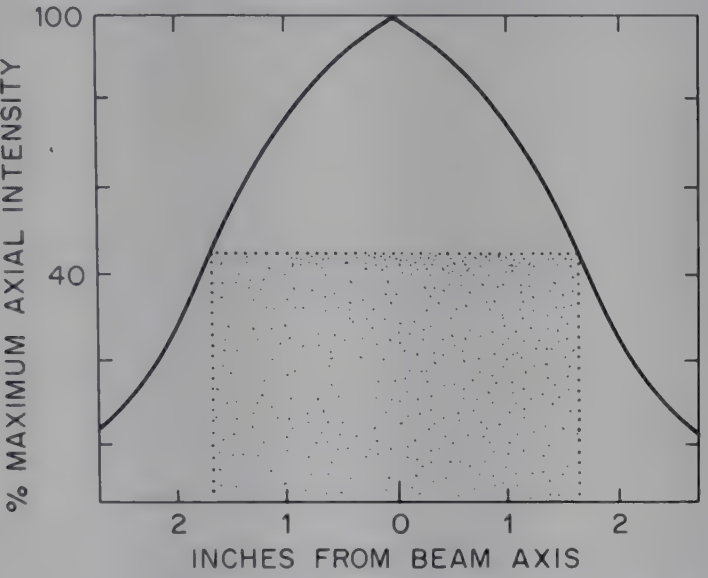
Go-No-Go Dosimeters.—With the establishment of the dose rate of a radiation source and the dose absorbed in a substrate, it is possible to have available workable monitoring devices which are called Go-No-Go dosimeters. Each unit container of substrate being irradiated should carry such a device. These production control devices may be as simple as colored markings on a container or colored plastic tapes. Here again color changes are related to dose received. Although the Go-No-Go dosimeters are not as precise as the primary and secondary standards, these inexpensive controls are extremely useful in positively indicating that a radiation treatment was given to the unit package of the substrate.

When this, plus the information obtained with more precise dosimeters which are monitoring the radiation source itself, are coupled, there is little opportunity for error in establishing that a given unit has received at least the dose of radiation intended (Hanley 1954).

Other Dosimeters.—Any reproducible, measurable radiation effect can serve as a dosimeter. The number of different dosimeter systems already developed for a variety of purposes is constantly expanding. A number of additional types are discussed in reference.

Dose Distribution

As indicated in Chapter 6, the administration of a dose of radiation to a unit container must be controlled. The dose distribution within a packet and between packets is a significant factor in quality control. To irradiate a substrate successfully, it is necessary to have information on the spatial distribution of radiation emitted from a source as well as information on the energy absorbed from this flux by the irradiated material. As noted earlier, the energy absorbed by unit volume in unit time depends on the nature of the radiation and the composition of the particular material being irradiated. The substrate determines the extent to which the radiation is absorbed. It must be noted that the dosimeter problem is not solved by specifying the characteristics of the radiation installation.



Courtesy of High Voltage Engineering Corp.

FIG. 29. OPTIMUM DISTRIBUTION OF ELECTRON ENERGY ACROSS THREE-INCH CONTAINER WITH UNSCANNED BEAM

Gamma Source

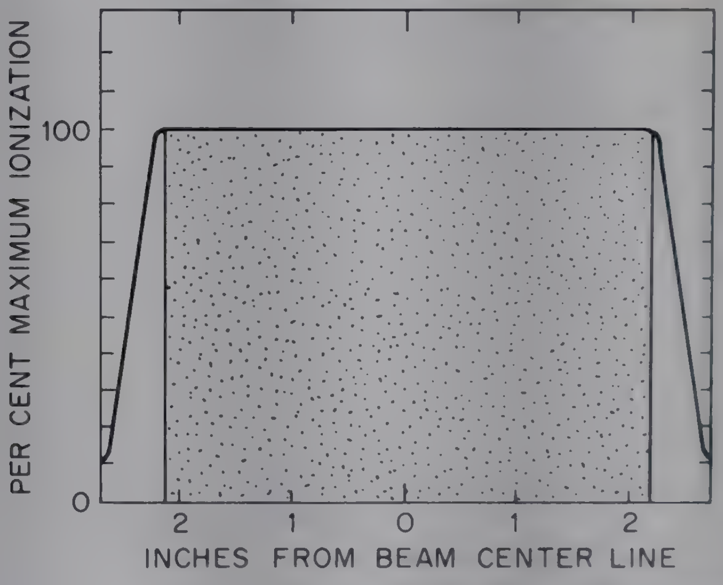
In gamma sources consisting of a circular array of fuel elements, the source configuration is ideally suited to a cylindrical container. Theoretical considerations by Herschman reveal that the dose distribution of gamma rays in cylindrical cans, exposed in a cylindrical source of infinite length, would conform to the following equation:

$$D = D_0 \left(1 + {}^{1}/_{10} (r/R)^2\right)$$

D_0 is the dose at the center of the can
D is the dose at some point along the equatorial plane
r is the radius of a point along the equatorial plane
R is the can radius

Dose measurements indicate that experimental values are in close agreement with theoretical results. Consideration is being given to the use of flat, parallel cobalt-60 plates as a gamma source in the United States. With such a geometry, rectangular packages would be most desirable.

Herschman studied the dose distribution pattern in 6 by 7 inch cylindrical tin coated steel cans, using cobalt glass dosimeters. Maps indicating the variation within stationary containers of unit density exposed to gamma radiation from fuel elements yielding a flux of 0.5 × 10^6 rads/hour were made. Isodose curves are shown in Fig. 28. At least a 10 per cent variation is encountered.



Courtesy of High Voltage Engineering Corp.

FIG. 30. DISTRIBUTION OF ELECTRON ACROSS 5-INCH CYLINDRICAL CONTAINER USING SCANNING BEAM

Electron Source

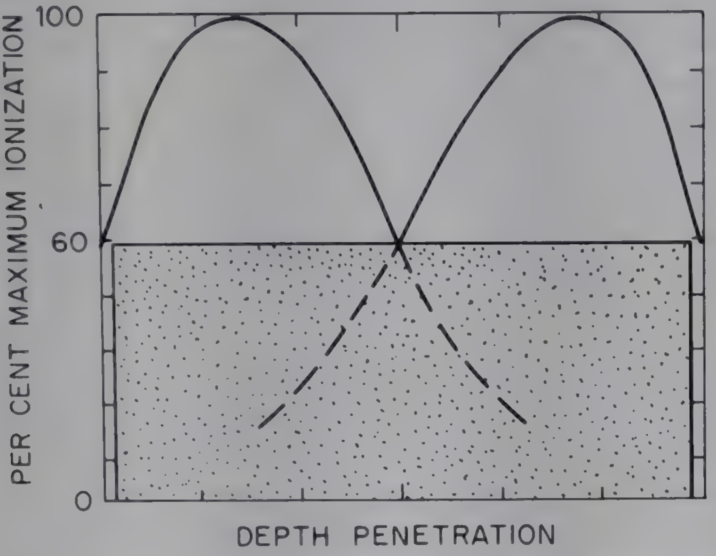
In the case of irradiation with high energy electrons, the electrons have a maximum range for a given energy calculated on the basis of the following equation, for electrons with energy greater than 0.8 Mev (Hine and Brownell 1956):

$$R_{\text{max.}} = \frac{0.542E - 0.133}{d}$$

$R_{\text{max.}}$ is the maximum range in centimeters
E is the energy of electrons in Mev
d is the density of irradiated material in gm./cm.^{3}

The above equation cannot be used directly since it describes a distribution of dose from 0 to 100 per cent. Therefore, a value is used known as the "effective range" which is equal to $^3/_5 R_{\text{max.}}$.

With an unscanned electron beam, the distribution of energy ranges from a maximum on the beam axis to decreasing values the further the object is from the beam axis. This is shown in Fig. 29. It is possible to broaden the effective area of use by scanning the beam (Fig. 30). With a scanned beam of electrons, uniformity of treatment is optimized.



Courtesy of U.S. Army Quartermaster Corps

FIG. 31. BOMBARDMENT OF CONTAINER FROM TWO SIDES

Improving the geometry of use without increasing the electron energies

Another means of improving the effective use or increasing the geometry of use without increasing the electron energies is to bombard a target material from two or more sides. Such a pattern is shown in Fig. 31.

Dosimetry and Health Physics

Personnel protection in any radiation installation also involves many dosimetry problems. In contrast to the radiation doses applied to process materials, which are in the order of millions of rads, the dose measurement range appropriate to the personnel protection problem lies in the range of milliroentgen, approximately equal to a millirad. The accumulation of one roentgen is a serious problem to a human.

Tables 58 and 59 (pps. 317, 318) show the type of effects on humans which are to be expected for varying radiation doses. For example 400 r usually will kill, and there may be radiation sickness from as little as 20 r. In view of this, the permissible radiation doses for workers in a site have been set after careful study at the low figure of 300 milliroentgen per week.

The dosimetry techniques appropriate to this type of problem are quite different from those being discussed in this chapter and will be taken up in Chapter 21.

BIBLIOGRAPHY

Ártandi, C., and Stonehill, A. A. 1958. Polyvinyl chloride—new high level dosimeter. Nucleonics *16,* No. 5, 118–120.

Davison, S., Goldblith, S. A., and Proctor, B. E. 1956. Glass dosimetry. Nucleonics, *14,* No. 1, 34–36.

Dudley, R. A. 1954. Photographic detection and dosimetry of beta rays. Nucleonics, *12,* No. 5, 24–26.

Feng, P. Y. 1958. Polymer Degradation—wide-range dosimeter, Nucleonics. *16,* No. 10. 114–116.

Fowler, J. F., and Day, M. J. 1955. High dose measurement by optical absorption, Nucleonics. *13,* No. 12, 52–54.

Fricke, H., and Morse, S. 1928. The chemical action of Roentgen rays on dilute ferrous sulphate solutions as a measure of dose. Am. J. Roentgenol. Radium Therapy, *18,* 430–431.

Gamma measurement, summary of 1958 Geneva Conference. Nculeonics, *16,* No. 9, 84, 1958.

Hart, E. J., and Gordon, S. 1954. Gas evolution dosimetry of high gamma neutron fluxes. Nucleonics, *12,* No. 4, 40–42.

Hanley, E. J. 1954. Gamma ray dosimetry with cellophane-dye systems. Nucleonics, *12,* No. 9, 62–64.

Harlan, J. T., and Hart, E. J. 1959. Ceric dosimetry: accurate measurements at 10^8 rads. Nucleonics, *17,* No. 8, 102–104.

Herschman, A. 1957. Theoretical analysis of the dose distribution in a No. 10 can. QM. Food and Container Institute Report Nos. 32-57, 41-57, 36-57.

Hine, G. J., and Brownell, G. L. 1956. Radiation dosimetry, Academic Press, Inc., New York.

Kreidl, N. J., and Blair, G. E. 1956. A stream of megaroentgen glass dosimetry. Nucleonics, *14,* No. 1, 56–58.

Magill, R. J. 1954. Simplified film processing for radiation dosimetry. Nucleonics, *12,* No. 8, 43–45.

McLaughlin, W. L., and Ehrlich, M. 1954. Film badge dosimetry: how much fading occurs? Nucleonics, *12,* No. 8, 34–36.

MOODY, J. W., KENDALL, G. L., and WILLARDSON, R. K. 1958. Photovoltaic gamma ray dosimeter. Nucleonics, *16*, No. 10, 101–103.

NITKA, H. F., and JONES, D. P. 1957. Photographic method for megaroentgen dosimetry. Nucleonics, *15*, No. 10, 128–129.

Nucleonics Report. 1959. Measuring large radiation doses. Nucleonics, *17*, No. 10, 57–76.

O'BRIEN, K., SOLON, L. R., and BLATZ, H. 1956. A system of film-badge interpretation. Nucleonics, *14*, No. 10, 58–60.

Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, 1955, Vol. 14. General aspects of the use of radioactive isotopes: dosimetry. United Nations, New York.

SCHAFFERT, J. 1957. Portable, self-powered reader for DT-60 glass dosimeter. Nucleonics, *15*, No. 12, 60–62.

SCHUCH, R. L., HIEBERT, R. D., and HAYES, F. N. 1954. Photovoltaic scintillation detector for high dose rates. Nucleonics, *12*, No. 2, 16–18.

SCHULMAN, J. H., KLICK, C. C., and RABIN, H. 1955. Measuring high doses by absorption changes in glass. Nucleonics, *13*, No. 2, 30–32.

SPEARMAN, JR., K. R. 1956. Correcting Anthracene dosimeter readings for neutron response. Nucleonics, *14*, No. 11, 84–86.

TAIMUTY, S. J., FOWLE, L. H., and PETERSON, D. L. 1959. Ceric dosimetry for routine use for 10^5 to 10^7 rads. Nucleonics, *17*, No. 8, 103–107.

WEISS, J., ALLEN, A. O., and SCHWARTZ, H. A. 1955. Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Geneva, Vol. *14*, 179–181. United Nations, New York.

PART III

Interaction of Radiation With Living Organisms

Radiolysis of Water and Aqueous Solutions

Since the discovery of the chemical effects of radiation some decades ago, many studies have been undertaken on systems varying in complexity from the inert gases to proteins and enzymes. The literature in this field is well into the thousands for journal publications and research reports. Considering the vast number of chemicals, however, the surface has barely been scratched. In view of this, it is of interest to note that a considerable amount of the research effort has been devoted to a study of a single, simple substance—water. A reason for this appears to be the central importance of this substance—water—not only to man's way of life, but to life itself. All living material contains water, in many instances it is the most abundant single constituent. It is thus to be expected that the effects of radiation on this substance will play an important part in shaping the gross effects of radiation on living matter.

The radiolysis of water produces only two substances, hydrogen and hydrogen peroxide. One can thus write formally

$$H_2O\ (\text{liquid}) \xrightarrow{\text{radiation}} H_2(\text{gas}) + H_2O_2(\text{liquid})$$

The molecular events leading to the transformation of this one simple substance into two others are rather complex. Although one could write a balanced chemical equation such as

$$2H_2O \rightarrow H_2 + H_2O_2$$

this equation contains no information which contributes to understanding the way in which the products are formed or the fact that the amount of each of the products may depend on temperature, trace impurities, pH, the kind of radiation employed, and on the intensity of the radiation source. The sensitive dependence of radiation effects on factors such as the above is quite general and must be taken into consideration in each case.

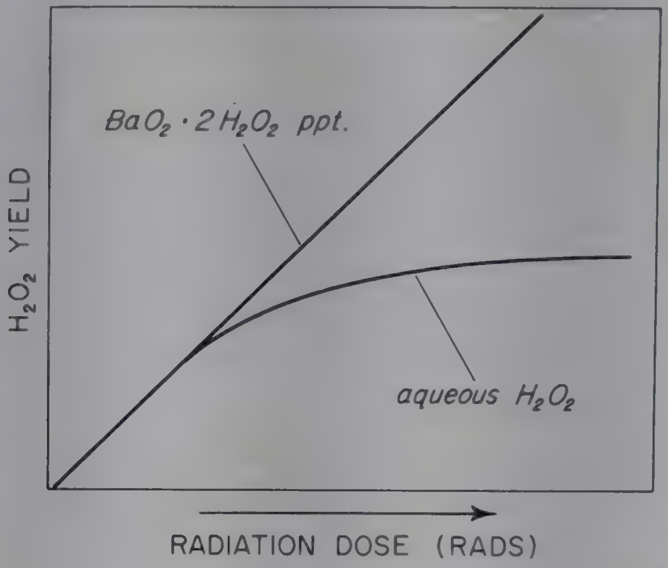
Many features of the radiolysis of water and aqueous solutions can be explained on the assumption that the primary active species[1] produced by radiation consist of hydrogen atoms and hydroxyl radicals,

$$H_2O \rightsquigarrow H\cdot + \cdot OH$$

first suggested by J. Weiss. The precise details of the mechanism of the formation of these species are still in dispute, particularly in connection with the fate of electrons knocked out of the molecule by radiation and the resulting spatial distribution of hydrogen atoms and hydroxyl radicals (Dainton 1959). Important as these differences are for a complete understanding of water radiolysis, they can be neglected for the purposes of the present discussion. These radicals are formed in close association with one another in the spurs of the radiation track. In each spur there are of the order of five or six such radical pairs. Following formation in the spurs, the radicals may recombine or diffuse away. Of the recombination reactions, one restores the original molecule and two others form hydrogen and hydrogen peroxide.

---

[1] See footnote p. 44, Chapter 4.



Courtesy of Veselovsky

FIG. 32. HYDROGEN PEROXIDE YIELD ALTERED BY INHIBITING BACK REACTION, PRECIPITATING HYDROGEN PEROXIDE AS IT IS FORMED

$$H\cdot + \cdot OH \rightarrow H_2O$$
$$H\cdot + H\cdot \rightarrow H_2$$
$$\cdot OH + \cdot OH \rightarrow H_2O_2$$

As the unrecombined radicals diffuse away they can react with hydrogen and hydrogen peroxide molecules which were formed at different sites according to the reactions

$$H\cdot + H_2O_2 \rightarrow H_2O + \cdot OH$$
$$\cdot OH + H_2 \rightarrow H_2O + H\cdot$$

These reactions are called back reactions because they restore the original material. In particular, as the concentration of hydrogen peroxide builds up during radiolysis, the rate of its destruction by hydrogen atoms increases until the rate of destruction equals the rate of formation. At this point, the hydrogen peroxide no longer builds up, but reaches a steady state concentration. This reaction scheme can be verified by adding barium oxide to the water which precipitates the hydrogen peroxide as fast as it is formed and interferes with the back reaction. It is found that the precipitate, which consists of $BaO_2 \cdot 2H_2O_2$, is formed at a constant rate. See Fig. 32 (Veselovsky *et al.* 1955).

Chemical Yields and G values

In the discussion to follow it will be useful to discuss the yields of radiation chemical reactions. A measure of the yield is the number of molecules produced or the number of radicals formed when a given amount of radiation energy has been absorbed by the irradiated material. The measure which has been adopted is the *G* value which is the number of molecules produced or radicals formed per 100 ev of absorbed energy. Generally speaking, except for polymerization and chain reactions, *G* values range roughly from 0.01 to 10. It is also convenient—and consistent with its definition—to use a *G* value as a measure of the number of molecules of starting material which are destroyed or transformed per 100 ev of absorbed radiation. In order to compute overall chemical yield or concentration from a *G* value, it is necessary to specify the total amount of energy absorbed per gram or the energy absorbed per unit volume, respectively. The latter are directly related to the radiation dose which is discussed in detail in Chapter 7.

The Cage Effect

As indicated in Chapter 5, an important factor in radiation chemical change is the ability of fragments of decomposing molecules to escape from the site of formation. If none of the fragments of a given molecule can escape, the excitation energy of the molecule may rapidly dissipate and the decomposition will not take place. In the case of water, the cage effect has been shown to be quite large. In gaseous water, approximately twelve molecules are decomposed per hundred electron volts of absorbed energy. In liquid water at room temperature, only about 4 to 5 molecules can be effectively decomposed with the same energy input. The difference is primarly due to two factors: the rate of energy dissipation is less in the gas phase due to the lower collision rate between molecules, and there is no serious obstacle to the escape of fragments from a decomposing molecule.

Scavenger Reactions

The elucidation of the great complexities of water radiolysis, which have only been indicated above, were made possible in part by a number of studies on the radiolysis of water to which was added a substance which could readily react with one or another of the radicals whose presence was suspected. Such substances are called scavengers.

An interesting example of their use is in connection with the problem of molecular hydrogen formation. Molecular hydrogen could be formed either directly by the decomposition of an excited water molecule, perhaps interacting with one or more neighboring ones.

$$H_2O^* + xH_2O \rightarrow H_2 + H_2O_2 + (x-1)\, H_2O$$

or, as indicated above, by the formation first of a hydrogen atom which later combines with another hydrogen atom to form H_2. If the latter reaction is the principal mechanism, it is expected that addition of a substance which readily reacts with hydrogen atoms would reduce the amount of molecular hydrogen formed. Experiments with solutions containing a high concentration of calcium nitrate, in which the nitrate ions readily react with hydrogen atoms, indicate that virtually no hydrogen gas is formed. This strongly supports the great predominance of the atomic mechanism (Hart 1959).

A further example of scavenging action is the effect on hydrogen peroxide formation of adding hydrogen atom scavengers. The addition of acrylamide in even very small concentration interferes with the back reaction between hydrogen atoms and hydrogen peroxide and thus increases the yield of the latter. Similar effects are observed in the presence of excess oxygen which undergoes the reaction

$$H\cdot + O_2 \rightarrow HO_2\cdot$$

The $HO_2\cdot$ radical cannot destroy hydrogen peroxide, and the yield of the latter increases.

Additional Radical Species

In addition to the radicals $H^\cdot$ and $^\cdot OH$ there is growing evidence that a number of other active species appear to be formed by secondary reactions in irradiated water. These suggested species include $HO_2^\cdot$, H_2^+, O^- and perhaps solvated electrons (Dainton 1959). Solvated electrons are analogous to solvated protons. It is known that the hydrogen ion H^+ exists in water in association with a water molecule, forming the hydronium ion H_3O^+. The solvated electron, a negative charge, is presumably associated also with one or more water molecules.

On the basis of oxygen formation in air-free irradiated water containing $FeSO_4$ and $CuCO_4$ it has been suggested that $HO_2^\cdot$ radicals are formed to some extent by the secondary reaction

$$H_2O_2 + \cdot OH \rightarrow H_2O + HO_2\cdot$$

The evidence for H_2^+ is again based on indirect considerations of hydrogen formation in air-free $FeSO_4$ solutions. The species O^- is postulated on the basis of isotopic exchange reactions with oxygen gas containing the isotope O^{18} and the dependence of the reaction on the acidity of the solution (pH) which affects the equilibrium

$$\cdot OH \rightarrow O^- + H^+\cdot$$

The evidence for the solvated electron is very indirect. An excellent comprehensive discussion of the general kinetics of water radiolysis has been given by Allen and Schwartz (1958).

Linear Energy Transfer Effects

In the discussion on physical effects of radiation it was pointed out that the different types of radiation, e.g., fast electrons and atomic radiation such as particles and fission fragments have significantly different penetration ranges in any given substance. Whereas electrons of several Mev energy can penetrate into liquid water for several centimeters, the penetration of alpha particles of the same energy is only a few millimeters and still less for the much heavier fission fragments. As a consequence of this, the spatial distribution of excitation and ionization is considerably different for electrons and atomic radiation. Whereas in the case of fast electrons the active species are distributed in distinct spurs containing on the order of six radical pairs, the alpha particle track consists of a continuous column of active species. These differences in spatial distribution affect the competition between reactions involving recombination of the active species and the reactions in which an active species diffuses away from the site of formation and subsequently reacts with a solute molecule. These effects have been demonstrated by means of scavengers whose reactions give a measure of the hydrogen atoms and hydroxyl radicals available for solute molecule reactions (Hart 1959). With these techniques it has been found that the overall decomposition yield of water is very nearly the same for gamma rays, protons, deuterons, alpha particles, and fission fragments. The G value for water decomposition decreases only from 4.5 for gamma rays to 3.6 for fission fragments. In contrast, however, the yield of radicals available for solute reactions decreases much more markedly, dropping from 6.0 to essentially zero. If, as seems to be the case, hydrogen and hydrogen peroxide formation involves radical intermediates to the exclusion of direct formation of molecular products, then the yield or G value for available radicals should be at most, twice the G value for water decomposition.

Oxidation-Reduction Reactions in Aqueous Solutions

If one recalls the classic definition of oxidation and reduction as the loss and gain of electrons, respectively, it is apparent that the radicals produced in water can function as oxidizing and reducing agents. A hydrogen atom can be a reducing agent or electron donor

$$H\cdot + H_2O \rightarrow H_3O^+ + e$$

and a hydroxyl radical can be an electron acceptor or oxidizing agent.

$$\cdot OH + e \rightarrow OH^-$$

A great variety of radiation-induced oxidation and reduction reactions in aqueous solutions have in fact been found to occur involving comparatively simple ions such as iodide ion I^- ferrous ion Fe^{++}, polyatomic ions such as nitrite and nitrate and organic molecules such as alcohols, benzene, aldehydes, and complex dyes.

The oxidation of ferrous ions by radiation is of particular interest in radiation technology since this reaction forms the basis of one of the most widely used and widely studied chemical dosimeters. In the discussion to follow, the particulars of the chemical transformation apply, of course, to the oxidation of ferrous ion. However, the thread of the argument runs fairly generally through many other instances of radiation induced oxidations and reductions in aqueous solutions.

In an acidified aerated aqueous solution of a ferrous compound the following processes occur. Irradiated water yields hydrogen atoms and hydroxyl radicals. The hydroxyl radical can oxidize a ferrous ion and each hydrogen atom can also lead to the oxidation of three more ferrous ions by the steps:

$$\begin{gathered}
Fe^{++} + \cdot OH \rightarrow Fe^{+++} + OH^- \\
H\cdot + O_2 \rightarrow HO_2\cdot \\
Fe^{++} + HO_2\cdot + H^+ \rightarrow H_2O_2 + Fe^{+++} \\
Fe^{++} + H_2O_2 \rightarrow Fe^{+++} + \cdot OH + OH^- \\
Fe^{++} + \cdot OH \rightarrow Fe^{+++} + OH^-
\end{gathered}$$

In these reactions a low pH or high concentration of H^+ (or H_3O^+) favors these reactions. Any reaction resulting in the formation of OH^- will be enhanced because the OH^- is neutralized by H^+. The overall consequence of this set of reactions is that for every water molecule decomposed, a total of four ferrous ions are oxidized provided all the radicals are available for reaction with solute molecules. In this connection it is interesting to note that with gamma radiation the *G* value for ferrous ion oxidation has been established to be 15.6 which is only slightly less than four times the *G* value for water decomposition, 4.5. Further, the *G* value for ferrous ion oxidation decreases progressively for heavier particles such as deuterons, alpha particles, and fission fragments to a low of about 5.4. This is in qualitative agreement with the discussion on linear energy transfer effects.

If an air saturated solution of ferrous ion is irradiated for a lengthy period of time, the rate of increase of ferric ion becomes progressively less since the oxygen is gradually consumed.

$$H\cdot + O_2 \rightarrow HO_2\cdot$$

This decreases the rate of the reaction, and permits more effective competition of the back reaction in which the hydrogen atom plays the more usual role of reducing agent.

$$H\cdot + Fe^{+++} \rightarrow H^+ + Fe^{++}$$

In alkaline solution the radical $HO_2\cdot$ can also act as a reducing agent with the liberation of oxygen.

$$Fe^{+++} + HO_2\cdot \rightarrow Fe^{++} + H^+ + O_2$$

Thus, in the absence of oxygen and/or in alkaline solution the yield of ferric ion is much reduced.

Another reaction of interest in connection with dosimetry is the reduction of ceric ion, Ce^{++++}, to cerous ion, Ce^{+++}. The reaction involved is

$$Ce^{++++} + H\cdot \rightarrow H^+ + Ce^{+++}$$

which is offset by the reverse reaction.

$$Ce^{+++} + \cdot OH \rightarrow Ce^{++++} + OH^-$$

In solutions of ceric salts the net G value for reduction is about 2.5 while on addition of a scavenger (such as thallous ion) for the oxidizing hydroxyl radicals, the reduction G value increases to as much as 7.9. The low G value for ceric ion reduction as compared to ferrous oxidation ($G = 15.6$) provides, with appropriate analytical methods, a dosimeter useful in a higher dose range than for the ferrous sulfate dosimeter.

The two systems discussed above serve only to illustrate some of the general features of the oxidation and reduction reactions which can occur in irradiated aqueous solutions. Numerous examples have been studied in greater or lesser detail. These include the oxidation of iodide ion via the reaction $I^- + \cdot OH \rightarrow I\cdot + OH^-$ reduction of nitrate ion by hydrogen atoms nitric oxide $NO_2^- + H\cdot \rightarrow NO + OH^-$ and reduction of cupric ions by hydroperoxyl radical $HO_2\cdot + Cu^{++} \rightarrow Cu^+ + H^+ + O_2$. Many examples are given by Hart (1959). The general result is that the overall effects of irradiation of aqueous solutions are largely explicable in terms of "conventional" reaction mechanisms with the additional complication that the concentration of the reacting species may be non-uniform and time-dependent. It is to be expected, and will be shown in the next chapters, that a great variety of oxidation and reduction reactions can occur in aqueous solutions of organic compounds. These reactions are for the most part understandable in terms of the effects of $H\cdot$, $\cdot OH$ and $HO_2\cdot$ radicals on the organic material.

BIBLIOGRAPHY

Allen, A. O. 1952. Mechanism of decomposition of water by ionizing radiations. Faraday Society Discussion No. 12, 79–87.

Allen, A. O., and Schwartz, H. A. 1958. Decomposition of water under high energy radiation. Geneva Conference *29*, 30–37.

Burton, M. 1951. An introduction to radiation chemistry. J. Chem. Educ. *28*, 404–413.

Burton, M. 1952. Elementary chemical processes in radiobiological reactions. Symposium on Radiobiology, J. J. Nickson, Ed., John Wiley and Sons. 117–138.

Dainton, F. S. 1959. Chemical effects of radiation. Radiation Research. Supplement No. 1, 1–25.

Dewhurst, H. A., Samuel, A. H., and Magee, J. L. 1954. A theoretical survey of the radiation chemistry of water and aqueous solutions. Radiation Research *1*, 62–84.

Fricke, H., and Morse, S. 1929. The action of X-rays on $FeSO_4$ solutions. Phil. Mag. *7*, 129–34.

Hart, E. J. 1959. Development of the radiation chemistry of aqueous solutions. J. Chem. Educ. *36*, 266–272.

Kuppermann, A. 1959. Theoretical foundations of radiation chemistry. J. Chem. Educ. *36*, 279–285.

Laidler, K. J. 1954. Elementary processes in the radiation chemistry of water. J. Chem. Phys. *22*, 1740–1745.

Lassettre, E. N. 1959. Collision cross section studies on molecular gases and the dissociation of oxygen and water. Radiation Research, Supplement No. 1, 530–546.

Scholes, F., and Weiss, J. 1959. Oxygen effects and formation of peroxides in aqueous solutions. Radiation Research, Supplement No. 1, 177–189.

Veselovsky, V. I., Miller, N. B., and Shub, D. M. 1955. Investigation of the laws of radiochemical homogeneous and heterogeneous formation of hydrogen peroxide. Symposium on Radiation Chemistry, Acad. of Sci., USSR. Moscow. Part 1, 35–45. In English translation by Consultants Bureau, Inc., New York.

Walling, C. 1957. Free Radicals in Solution. John Wiley and Sons, Inc., New York.

CHAPTER 9

Radiation Effects on Organic Compounds

Introduction

The radiation chemistry of organic compounds covers a field potentially as vast as the "conventional" chemistry of organic compounds. Already the number of publications in this field is well into the thousands. And yet the field must be regarded as being still in its infancy. Many interesting and useful types of chemical changes induced by radiation have been demonstrated on a variety of organic chemical systems. But our detailed knowledge is all too scant. The work to date has, however, led to the conclusion that there are large areas of overlap and parallelism between radiation induced and "conventional" chemical change, and the areas of dissimilarity are becoming defined with ever-increasing clarity and precision.

Given the experimental result that irradiation of a particular pure substance produces definite amounts of a number of products, it is desirable to account both for the formation of the various products and, further, to account for the quantity of these products. It is in connection with the first, the qualitative question, that much of our knowledge of "conventional" organic chemistry is useful. Many qualitative aspects of organic radiation chemistry are explicable in terms of free radical reactions which are analogous if not identical with the well established free radical reactions known in organic chemistry. Further, the existence of such free radicals in irradiated systems has been amply demonstrated by both chemical and physical techniques. One point of departure from conventional chemistry is the non-uniform spatial distribution of the radicals and other active species. This has already been discussed in the preceding chapter. This fact alone poses serious difficulties in any quantitative discussion of the course of the various chemical reactions. In addition, information concerning the precise nature and abundance of the active species initially formed by irradiation is still very incomplete. Experimental work on this problem has in some cases led to reasonably complete and accurate information on the nature and amount of various free radicals formed by radiation interaction in some organic liquids. However, the fundamental physical and chemical factors leading to this set of active species are still far from clear. There is abundant evidence that the composition of this set is far from random, but so far it has received no quantitative theoretical accounting. Thus, being faced with a complex set of reactions initiated by a variety of active species, we must at present be restricted to qualitative considerations of mechanism.

The present chapter presents a brief survey of the radiation chemistry of non-polymeric organic compounds in the order hydrocarbons, alcohols, ethers, aldehydes and ketones, acids, amino acids, simple carbohydrates, fats, vitamins and steroids, and pigments. Where available, information is included on the radiation chemistry of the compounds in pure form, in mixtures, in the presence of oxygen, in aqueous solution and in natural surroundings, such as vitamins irradiated while incorporated in a food substance. It will be seen that the presence of other substances frequently has a significant effect on the result of the irradiation.

Hydrocarbons

The irradiation of saturated aliphatic hydrocarbons produces two significant gaseous products, namely hydrogen and small amounts of methane. Traces of two, three, and four carbon products have also been identified in the gas phase, consisting of both saturated and unsaturated molecules. The liquid phase products include olefins having the same number of carbon atoms as the starting material, a variety of isomeric alkanes of higher molecular weight up to twice that of the starting material, and some high molecular weight crosslinked polymers (Dewhurst 1957).

The hydrogen gas can be formed in two ways. First, by the formation of a hydrogen atom and alkyl radical, followed by the abstraction of another hydrogen atom from another molecule. Second, this product may be formed directly by the decomposition of an excited molecule into an olefin and a hydrogen molecule. The two paths may be illustrated for *n*-hexane by the equations:

$$C_6H_{14}^* \rightarrow C_6H_{13}\cdot + H\cdot \qquad \text{(Ia)}$$

$$C_6H_{14} + H\cdot \rightarrow C_6H_{13}\cdot + H_2 \qquad \text{(Ib)}$$

and

$$C_6H_{14}^* \rightarrow C_6H_{12} + H_2 \qquad \text{(II)}$$

Subsequent reaction between the two hexyl radicals will form either dodecane, $C_{12}H_{26}$, by a recombination reaction or hexane and hexene by disproportionation. Depending on the structure of the radicals (or the location of the ruptured bond) a variety of isomers is obtained. A possible process for polymer formation consists of the formation of a $C_{12}H_{25}$ radical by hydrogen abstraction from the dimer $C_{12}H_{26}$ by a hydrogen atom, followed by recombination with another hexyl radical, and so on.

The gaseous hydrocarbon products as well as the liquid products containing seven to eleven carbon atoms are formed by reactions all of which initially involve carbon-carbon bond rupture of an excited hexane molecule. The possible radical pairs that can be formed include methyl-pentyl, ethyl-butyl, and propyl-propyl. The overwhelming predominance of methane among the gaseous hydrocarbon end products points to a predominance of the first type of cleavage. If carbon-carbon bond energies were the sole factor determining the probability of cleavage, the first process would have the smallest probability among the three. However, an important factor in the liquid state is the problem of escape of the radical fragments from one another. It is not sufficient that the bond be "broken"; it is necessary that one of the radicals bodily move out of the liquid cage to another position in order that immediate recombination does not reverse the process. In this problem of escape it is clear that the smaller radical is favored, and it would appear that this step controls the reactions. Following the separation of the radical pair the various products can again be formed by abstraction, recombination and disproportionation reactions. There is abundant evidence that the probability of bond rupture of the excited alkane molecules is definitely not random.

TABLE 13

ALKYL IODIDES FORMED IN IRRADIATED BUTANE-IODINE SOLUTIONS

Compound	*G* Value
CH_3I	0.47
C_2H_5I	0.94
n-C_3H_7I	0.09
i-C_3H_7I	0.04
n-C_4H_9I	1.04
Sec-C_4H_9I	2.14
CH_2I_2	0.64
Residual Iodides	0.35
Total Iodide	5.71

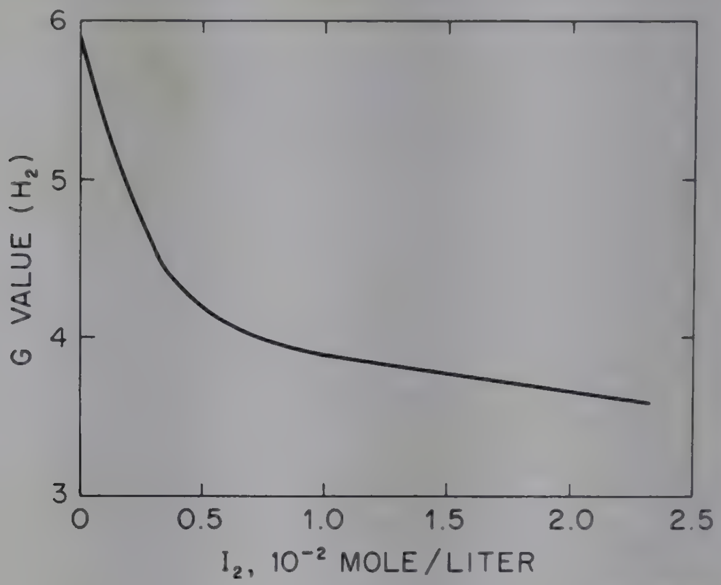
From McCauley & Schuler (1957)

Evidence for the formation of radicals in irradiated alkanes has been obtained by various techniques. One of the most important of these is the iodine scavenger method. It has been found that iodine molecules react extremely rapidly with organic free radicals, especially aliphatic radicals, according to the following equation. The resultant iodine atom

$$R\cdot + I_2 \rightarrow RI + I\cdot$$

will not form any organic radical by an abstraction process (Schuler 1958). Thus, if the reaction rate is rapid enough compared to the other types of radical reactions discussed above, the amount of various alkyl iodides formed is a measure of the quantity of organic radicals formed by irradiation, and an analysis of the nature of the iodides gives information on the types of free radicals formed. Some results of McCauley and Schuler (1957) on the application of this technique to the radiolysis of liquid butane are shown in Table 13. These results were obtained with 0.0024 molar iodine concentration. The fact that reproducible iodide yields are frequently obtained at very low iodide concentrations, independent of actual concentration, gives strong support for the validity of the method. Other types of scavenger techniques have been discussed by Prévot-Bernas and co-workers (1952).

The question of the relative importance of atomic and molecular mechanisms for formation of hydrogen gas has also been studied by iodine scavenger techniques. It is known that iodine is a scavenger for hydrogen atoms, readily forming hydrogen iodide and interfering with the formation of H_2 gas by the atomic mechanisms. It has been found that the addition of very small quantities of iodine, about 5×10^{-3} molar, reduces the hydrogen yield from a *G* value[1] of about 6 to a value of about 4 (Burton *et al.* 1957). Further six fold increase in iodine produces only a small additional decrease to about 3.5 (see Fig. 33). This type of study suggests the presence of both atomic and molecular mechanisms. In the case of benzene, the radiolytic production of hydrogen appears to be principally a molecular mechanism.



Courtesy of Burton

FIG. 33. EFFECT OF IODINE ON H_2 YIELD FROM CYCLOHEXANE

The processes discussed up to this point have all been either molecular or free radical processes. Another class of reactions, ion-molecule reactions, was discussed in Chapter 4. These reactions have very rapid rates compared to neutral free radical reactions. Further, it is clear that considerable numbers of ions are formed by the interaction of radiation with molecules. Nevertheless, the main features of radiolysis of organic liquids appear explicable in terms of neutral free radical mechanisms. The relative importance of ion-molecule reactions in liquids is at present not clearly defined. Up to the present the most convincing demonstrations of ion molecule reactions in liquids are in connection with polymerization reactions. For example, Chang, Yang and Wagner (1959) found that the radiation-induced dimerization of 1-olefins led to products that could be explained only by assuming ion-molecule reactions and not free radical reactions. This type of reaction may be schematized as:

---

[1] Molecules decomposed per 100 electron volts of energy absorbed.

$$A^+ + A \rightarrow AA^+$$

In contrast, the types of reactions most frequently encountered are of the type:

$$A + B \rightarrow C + D$$

that is, reactions involving the formation of two products. If it is recalled that the colliding partners of the reaction are confined in a liquid cage, it is seen that the reaction will not be completed until one of the products leaves the cage. This step is a diffusion process. It is possible that in such diffusion-controlled processes, the average time required for a separation of products is long compared to the time during which an electron is separated from its positive ion in a liquid. This would mean that the ion would be neutralized before the reaction could go to completion. Another way of stating the argument is as follows. The number of collisions between a molecule or a fragment and its neighbors in a liquid is of the order of 10^{13} per second. On the other hand, in the process of diffusion away from a particular lattice position in the liquid to a neighboring empty position, the molecule or fragment undergoes hundreds or even thousands of such collisions before it is properly directed with sufficient energy, and its neighbors so disposed in space, that the molecule can squeeze through to a neighboring vacancy. Thus, although an ion-molecule reaction in the isolated state is effective at every collision, in the liquid many more collisions must occur before the products can separate. And the time an ejected electron spends away from its partner ion is probably not that long. Now the circumstances that make the olefin dimerization a possible ion-molecule process is that the reaction involves no escaping products; it is complete in one collison and could go on to produce higher polymers before the electron returns. In actual fact dimers, trimers and tetramers have been found in irradiated 1-hexene. The crucial importance of the return time of the electron suggests that the presence or addition of substances which could trap the electrons temporarily might have pronounced effects on the reactions following initial excitation and ionization.

A rather general feature of hydrocarbon radiolysis is the relative stability of aromatic hydrocarbons compared to saturated hydrocarbons. An example is shown in Table 14, adapted from the work of Manion and Burton (1952). It is seen that the total product *G* values for radiolysis products are markedly lower in the aromatic compounds. This stability of the aromatic compounds is attributed in part to the ability of the aromatic compounds to re-radiate electronic excitation energy instead of having it all converted into vibrational excitation which ruptures chemical bonds. This explanation is supported by studies by Burton and his collaborators (Burton, Chang *et al.* 1957) which actually find light emission in these compounds in the presence of high energy radiation.

TABLE 14

PRODUCT YIELDS FOR RADIOLYSIS OF SOME PURE LIQUIDS

Substance	*G* Value				
	H_2	CH_4	C_2 gases	Polymer	Total
Benzene	0.036	—	0.022	0.76	0.82
Toluene	0.13	0.0077	0.0043	0.92	1.06
Cyclohexane	5.7	0.09	0.21	1.7	7.7
Cyclohexene	1.2	0.019	0.142	12.4	13.8

When a comparatively radiation-resistant compound such as benzene is mixed with a more labile substance such as cyclohexane it is found that the former acts as a protective agent. For example, if about 20 per cent of benzene is added to cyclohexane the hydrogen yield is reduced by about 80 per cent, far lower than expected from weighted average hydrogen yields for the pure substance. This protective effect is due in part to energy transfer from excited cyclohexane to benzene which can then reradiate it without undergoing chemical change. It may also be due in part to scavenging of hydrogen atoms by the benzene. This problem has been studied principally by Burton and his collaborators (Burton, Chang *et al.* 1957). In other cases such as gaseous hydrocarbon–rare gas mixtures, sensitizing effects are found, the rare gas transforming its excitation and charge to the hydrocarbon which then undergoes chemical reaction.

The principal reaction induced in unsaturated hydrocarbons by radiation is polymerization, both in the gaseous state and in the liquid state or in solution. The two possible mechanisms are free radical and ionic polymerization. The classic study in this field is the original work of Lind (Lind *et al.* 1925) on the α-particle irradiation of acetylene. The two products of the radiolysis were benzene and an insoluble waxy polymer which was called cuprene. The mechanism of this reaction is still not fully elucidated but it is believed that ions and excited acetylene molecules play a significant part (Dorfman and Shipko 1955). In recent years, the subject of radiation-induced polymerization has become a major field for research and development. It has been found that essentially all substances which can be polymerized by free radical initiators can be polymerized by high energy radiation. This is due to the formation of free radicals by the radiation. The fact that no chemical initiators, such as peroxide need be added to the monomer to induce polymerization may be of advantage in the radiation induced reaction. Much work has been done in this field, and it has a voluminous literature.

It is well known that at high temperatures hydrocarbons can be oxidized to a variety of products including alcohols, aldehydes, ketones, acids, peroxides and hydroperoxides (Semenov 1958). In many instances, these reactions involve a free radical mechanism of which the first step is the formation of a peroxyl radical from an organic radical and oxygen:

$$R\cdot + O_2 \rightarrow ROO\cdot$$

The peroxyl radical may now initiate a chain reaction by generating another organic radical, itself forming a hydroperoxide:

$$ROO\cdot + RH \rightarrow ROOH + R\cdot$$

Recombination of the peroxyl radical with another organic radical leads to the formation of a peroxide:

$$ROO\cdot + R\cdot \rightarrow ROOR$$

and a series of further reactions of the hydroperoxide and the peroxide leads to the formation of the other oxidation products, alcohols, aldehydes, ketones and acids. The first step of these subsequent reactions is the cleavage of the oxygen-oxygen bond of the peroxides forming oxy-radicals:

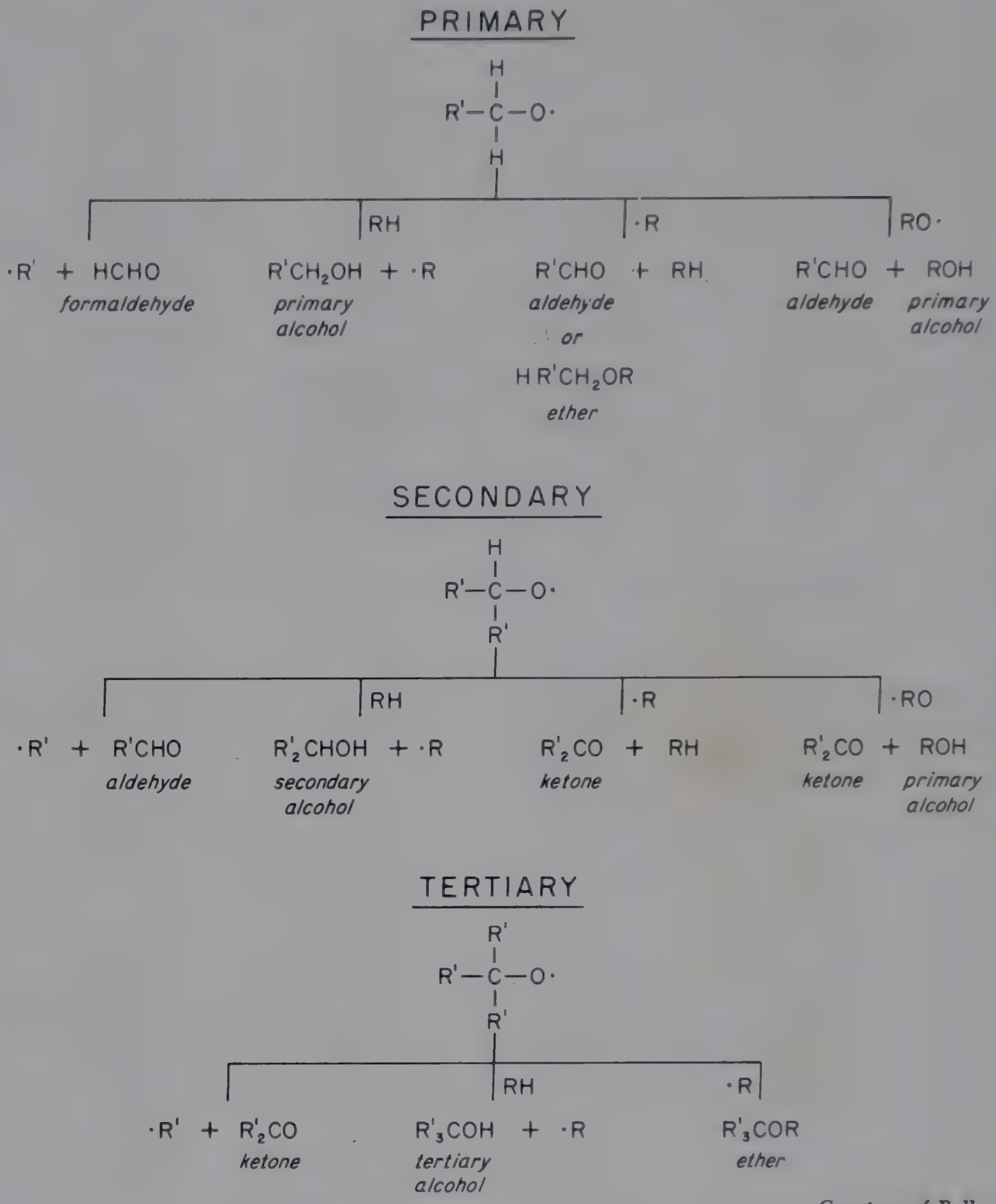
$$ROOR \rightarrow RO\cdot + RO\cdot$$

$$ROOH \rightarrow RO\cdot + \cdot OH$$

The further steps leading to the other oxidation products have been neatly summarized by Bell and collaborators (1953) in the reaction schemes for primary, secondary and tertiary oxy radicals shown in Fig. 34. Further oxidation leads to acids, as will be shown below. The hydroxyl radical forms water and a radical by hydrogen abstraction:

$$\cdot OH + RH \rightarrow HOH + R\cdot$$

This lengthy sequence of events is the key to the understanding of the results of radiation-induced oxidation. It was noted earlier that free radicals were formed among the primary products of hydrocarbon radiol-


PRIMARY
R'—C(H)(H)—O·
RH
·R
RO·
·R' + HCHO
formaldehyde
R'CH₂OH + ·R
primary alcohol
R'CHO + RH
aldehyde
or
HR'CH₂OR
ether
R'CHO + ROH
aldehyde
primary alcohol
SECONDARY
R'—C(H)(R')—O·
RH
·R
·RO
·R' + R'CHO
aldehyde
R'₂CHOH + ·R
secondary alcohol
R'₂CO + RH
ketone
R'₂CO + ROH
ketone
primary alcohol
TERTIARY
R'—C(R')(R')—O·
RH
·R
·R' + R'₂CO
ketone
R'₃COH + ·R
tertiary alcohol
R'₃COR
ether

Courtesy of Bell

FIG. 34. REACTIONS OF ALKOXY RADICALS

ysis. Further, the iodine scavenger experiments on butane, cited above, provide information on the relative amounts of the different types of radicals formed. If now a radiolysis is performed, for example, on a butane-oxygen mixture, the radicals will combine with oxygen to form peroxy radicals and the series of reactions given above in schematic form will ensue. It is clear that the variety of radicals formed by radiation will in general lead to an exceedingly complex mixture of oxidation products. However, the ability to produce radicals with radiation under any conditions of temperature and pressure offers greater freedom of choice of reaction conditions in order to enhance the formation of some reaction products and suppress others. This is notably the case in producing hydroperoxides and peroxides.

Alcohols

The principal gaseous product formed in the irradiation of pure alcohol is hydrogen. There are also smaller amounts of methane and carbon monoxide, but no carbon dioxide. Also found are quantities of saturated and unsaturated hydrocarbons having the same or a lesser number of carbon atoms, up to four carbons (the higher ones are liquid). The range of G values for the first three products, based on experiments with alcohols up to *n*-decyl alcohol (McDonnell and Newton 1954) are:

Product	G value
Hydrogen	1.24–3.59
Methane	0.018–1.14
CO	0.044–0.23

The principal liquid phase products include water, aldehydes, ketones, glycols and only traces of acid. The range of G values found is:

Product	G value
Water	0.31–1.00
Aldehyde	0.7–3.07
Ketone	0.00–2.78
Glycol	0.1–1.6
Acid	0.003–0.012

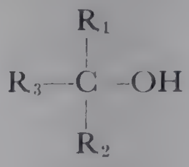
Trace quantities ($G < 0.1$) of other materials found include ethers and peroxides. The total yield of products corresponds to a decomposition of 7 to 10 alcohol molecules decomposed per 100 electron volts of energy input ($G = 7–10$).

The complexity of the reactions is illustrated by the fact that in irradiated ethanol a total of some 23 separate products have been identified. The nature and amount of all these products has yet to be quantitatively explained. However, some of the major features can be understood in terms of radical mechanisms. The formation of hydrogen, again, can proceed by atomic and molecular mechanisms. Indirect evidence for the atomic mechanism is found in the formation of peroxides, ROOR, which imply the prior formation of alkoxy radicals:

$$\mathrm{ROH} \rightarrow \mathrm{RO\cdot} + \mathrm{H\cdot}$$

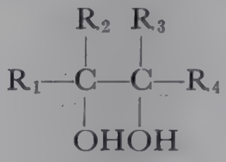
Further evidence for these alkoxy radicals is found in the oxidation products, aldehydes and ketones, which are formed via the alkoxy radical reaction scheme discussed above. Particularly noteworthy in this connection is that primary alkoxy radicals can form only aldehydes, not ketones, whereas secondary and tertiary alkoxy radicals can form both. It is found that the radiolysis of primary alcohols, methyl, ethyl, *n*-propyl, *n*-butyl, *i*-butyl, produces only aldehydes while ketones are found in irradiated *i*-propyl, *sec*-butyl and *t*-butyl alcohols. The presence of only traces of acid is consistent with the fact that molecular oxygen is required for the further oxidation of the aldehydes and ketones.

The weak bonds in the alcohol molecules are those connected to the hydroxyl-bearing carbon indicated in the diagram below.



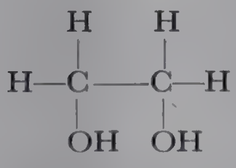
$$\begin{array}{ccc} & R_1 & \\ & | & \\ R_3\text{—} & C & \text{—}OH \\ & | & \\ & R_2 & \end{array}$$

The glycols found in the irradiated alcohols are predominantly the vicinal glycols in which the two hydroxyl groups are on adjacent carbon atoms:



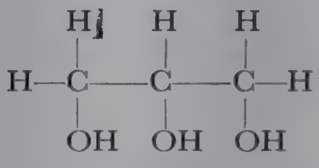
$$\begin{array}{cccc} & R_2 & R_3 & \\ & | & | & \\ R_1\text{—} & C\text{—} & C & \text{—}R_4 \\ & | & | & \\ & OH & OH & \end{array}$$

and result from recombinations of radicals in which C–H and C–R bonds have been ruptured on the hydroxyl-bearing carbon atom. Another study, on the self irradiation of C^{14} labeled methanol has found the formation of tri- and tetrahydroxy compounds as well as ethylene glycol (Skraba *et al.* 1953). The compounds are found in decreasing abundance in the order:



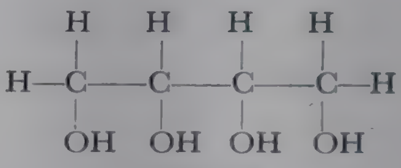
$$\begin{array}{cccc} & H & H & \\ & | & | & \\ H\text{—} & C\text{—} & C & \text{—}H \\ & | & | & \\ & OH & OH & \end{array}$$

ethylene glycol
1360



$$\begin{array}{ccccc} & H & H & H & \\ & | & | & | & \\ H\text{—} & C\text{—} & C\text{—} & C & \text{—}H \\ & | & | & | & \\ & OH & OH & OH & \end{array}$$

glycerol
14.9



$$\begin{array}{cccccc} & H & H & H & H & \\ & | & | & | & | & \\ H\text{—} & C\text{—} & C\text{—} & C\text{—} & C & \text{—}H \\ & | & | & | & | & \\ & OH & OH & OH & OH & \end{array}$$

erythritol
1.00

which are formed from further radiation-induced reaction of the dihydroxy compound.

It is also found that the methane yield in various alcohols depends on how many methyl groups are attached to the hydroxyl-bearing carbon. The methane is probably formed by formation of a methyl radical which subsequently abstracts a hydrogen atom (see Table 15).

In aqueous solution there do not appear to be any major changes in the radiolysis mechanisms. The hydrogen atom and hydroxyl radicals produced in water abstract a hydrogen from the dissolved alcohol, forming

TABLE 15

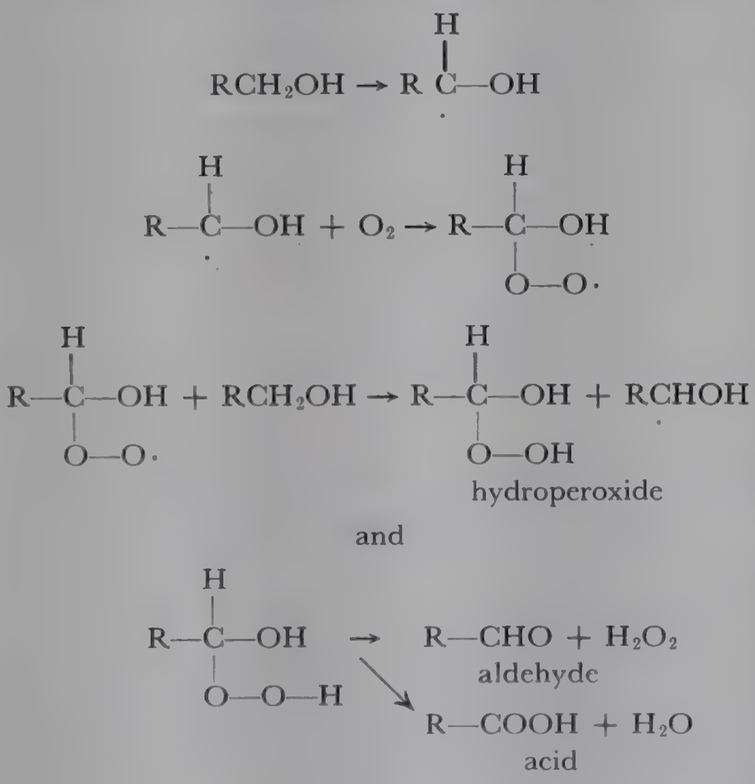
METHANE YIELDS IN IRRADIATED ALCOHOLS[1]

Methyl Groups on hydroxyl carbon	Compound	G_{CH_4}
None	Methanol	0.36
	n-Propanol	0.067
	n-Butanol	0.055
	i-Butanol	0.142
	n-Octanol	0.022
	n-Decanol	0.018
One	Ethanol	0.43
	sec-Butanol	0.37
Two	*i*-Propanol	1.14
Three	*t*-Butanol	1.60

[1] From McDonnell and Newton (1954).

hydrogen gas and water. Then the hydrogen yield is somewhat increased. No other pronounced changes appear.

In the presence of oxygen there are some changes in the radiolysis pattern. There is now an appreciable formation of acids and peroxides and an increase in hydrogen yield (Bakh *et al.* 1958). The peroxides are principally hydroperoxides. In the case of ethanol only hydrogen peroxide and oxyethyl hydroperoxide were found, $CH_3CHOH{-}OOH$. The reactions leading to the peroxides and the acids are as follows:



$$RCH_2OH \rightarrow R\overset{H}{\underset{\cdot}{C}}-OH$$

$$R-\overset{H}{\underset{\cdot}{C}}-OH + O_2 \rightarrow R-\overset{H}{\underset{O-O\cdot}{C}}-OH$$

$$R-\overset{H}{\underset{O-O\cdot}{C}}-OH + RCH_2OH \rightarrow \underset{\text{hydroperoxide}}{R-\overset{H}{\underset{O-OH}{C}}-OH} + R\underset{\cdot}{C}HOH$$

and

$$R-\overset{H}{\underset{O-O-H}{C}}-OH \begin{array}{l} \rightarrow \underset{\text{aldehyde}}{R-CHO} + H_2O_2 \\ \searrow \underset{\text{acid}}{R-COOH} + H_2O \end{array}$$

Ethers

The radiolysis of ethers is in many respects similar to the radiolysis of alcohols. The radiation-induced decomposition involves principally the rupture of the carbon-oxygen bond, followed by reaction of these radicals to form alcohols, aldehydes and ketones. Other reactions can be accounted for by the rupture of other bonds connected to the oxygen-bearing carbon. Among the products found are principally hydrogen and methane gas, and hydrocarbon resulting from the reaction

$$R_1\text{—}O\text{—}R_2 \rightarrow R_1\cdot + \cdot OR_2$$

$$R_1\cdot + R_1\text{—}O\text{—}R_2 \rightarrow R_1H + \text{oxy radical}$$

For example, the principal hydrocarbons found in various ethers are:

Ether	*G*-Hydrocarbon			
	CH_4	C_2H_6	C_3H_8	C_4H_{10}
ethyl	0.24	0.62	0.12	0.15
propyl	0.071	0.22	0.43	0.03
i-sopropyl	0.87	0.159	0.45	0.142
n-butyl	0.061	0.036	0.123	0.39

From Newton (1957).

A new reaction in this class of compounds is the rearrangement of an excited ether molecule to form an alcohol and an olefin

$$R_1\text{—}O\text{—}CH_2\text{—}CH_2\text{—}R_2 \rightarrow R_1OH + CH_2\text{=}CH\text{—}R_2$$

as indicated by the olefin yields below.

Ether	*G*-product		
	C_2H_4	C_3H_6	C_4H_6
Ethyl	1.04	0.01	0.15
Propyl	0.24	0.49	0.005
i-Propyl	0.33	1.14	0.008
n-Butyl	0.14	0.125	0.38

From Newton (1957).

A detailed study of the radiolysis of isopropyl ether shows that the yield of propylene is strongly dependent on the total energy input. Further, the addition of small amounts of acetaldehyde somewhat inhibits the formation of propane, propylene, isobutane and isobutylene. The effect is attributed to radical scavenging.

In aqueous solution, the irradiation of isopropyl ether produces acetone ($G = 3$) and isopropyl alcohol ($G = 1$), and it has been suggested (Bakh *et al.* 1959) that this reaction proceeds as

$$(CH_3)_2CH\text{—}O\text{—}CH(CH_3)_2 + H\cdot \text{ or } \cdot OH \rightarrow (CH_3)_2\underset{\cdot}{C}\text{—}O\text{—}CH(CH_3)_2 + H_2 \text{ or } H_2O$$

followed by a radical decomposition

$$(CH_3)_2\underset{\cdot}{C}\text{—}O\text{—}CH(CH_3)_2 \rightarrow \underset{\text{acetone}}{(CH_3)_2C{=}O} + \underset{\text{isopropyl radical}}{\cdot CH(CH_3)_2}$$

the isopropyl radical then combining with another hydroxyl radical to form the isopropyl alcohol. It is seen that this mechanism would remove hydroxyl radicals which otherwise would form hydrogen peroxide. Further, if excess hydrogen gas is dissolved in the solution, still no hydrogen peroxide and also no isopropyl alcohol are formed. In this case the hydrogen acts as a hydroxyl scavenger.

$$H_2 + \cdot OH \rightarrow H_2O + H\cdot$$

The yield of acetone is unchanged, which is reasonable since any hydroxyl that would abstract a hydrogen from the ether to start the process off is scavenged but replaced by a hydrogen atom which can attack the ether as well. In the presence of oxygen the acetone and isopropyl alcohol appear in equal amounts ($G = 1.40$) and hydrogen peroxide appears with a G value of 1.45. In this case hydrogen atoms combine with the oxygen to form $HO_2\cdot$ radicals which apparently cannot attack the ether but react to form hydrogen peroxide by the reaction

$$2\ HO_2\cdot \rightarrow H_2O_2 + O_2$$

The *G*-value of H_2O_2 suggests that not all hydrogen atoms formed in water attacked the ether in the absence of oxygen since the amount of hydrogen peroxide is about twice that expected from the decrease in acetone if all hydrogen atoms had attacked the ether. The irradiation of ethers in the presence of oxygen results in the formation of acids and peroxides as well as aldehydes and alcohols. In isopropyl ether the yield of alcohol increases by an amount equal to the yield of acid, both products being formed from a peroxide intermediate. This is in addition to the alcohol formed by mechanisms in pure ethers discussed earlier.

Aldehydes and Ketones

Compared to the alcohols the amount of information available on the radiation chemistry of aldehydes and ketones is exceedingly scarce. However, the photolysis of acetone with ultraviolet light is one of the classic reactions of photochemistry and has received much attention.

The products of photolysis of acetone include methane, ethane, carbon monoxide and biacetyl $(CH_3CO)_2$, whereas a recent study of acetone radiolysis reports the gaseous products hydrogen, methane and carbon monoxide. The absence of hydrogen in the photolysis products is due to the fact that the molecules do not receive enough energy to rupture a C–H bond. The radical processes are then

$$CH_3\ COCH_3 \rightarrow CH_3\cdot + CH_3CO\cdot$$

The methyl radical forms methane by hydrogen abstraction or ethane by recombination, while the acetyl radical forms biacetyl by combination or decomposes to carbon monoxide and a methyl radical.

In aqueous acetone solution no acids are formed, whereas aqueous acid solutions of acetaldehyde and propionaldehyde do form acids among their products. This difference may be due to the fact that hydroperoxyl radicals $HO_2\cdot$ formed from water can oxidize aldehydes but not ketones. It is to be expected that the irradiation of aldehydes or ketones in the presence of oxygen will produce the same types of oxidation products, including acids and peroxides, as discussed earlier (Bakh *et al.* 1958).

An interesting example of protective action, analogous to the benzene-cyclohexane mixture, is the depression of hydrogen yield in irradiated isopropanol by the addition of acetone (Strong and Burr 1959). It has been demonstrated here, with the use of isotopic substitution techniques, that the protective action of the acetone is due to its action as a hydrogen atom scavenger, and probably not due to energy transfer and dissipation.

Acids

No complete studies are available on the irradiation of pure carboxylic acids of low molecular weight. Partial studies on acetic acid indicate that the gaseous products include hydrogen ($G = 0.52$), carbon dioxide ($G = 4.0$), methane ($G = 1.38$), ethane ($G = 0.85$), and carbon monoxide ($G = 0.4$). It has been suggested that there are no non-volatile irradiation products in acetic acid (Garrison *et al.* 1955). Isotopic substitution studies indicate that both hydrogen and methane are produced primarily by free radical mechanisms such as (Burr 1957)

$$\mathrm{CH_3COOH} \xrightarrow{\text{radiolysis}} \cdot\mathrm{CH_2COOH} + \mathrm{H}\cdot$$

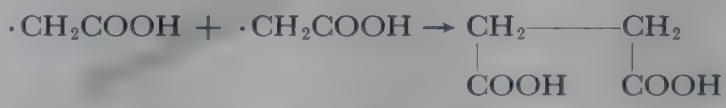
$$\mathrm{CH_3COOH} \xrightarrow{\text{radiolysis}} \mathrm{CH_3}\cdot + \cdot\mathrm{COOH}$$

followed by

$$\mathrm{H}\cdot + \mathrm{CH_3COOH} \rightarrow \mathrm{H_2} + \mathrm{CH_2COOH}$$

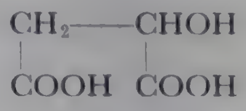
$$\mathrm{CH_3}\cdot + \mathrm{CH_3COOH} \rightarrow \mathrm{CH_4} + \cdot\mathrm{CH_2COOH}$$

In aqueous solution succinic acid is found, being formed by the recombination reaction



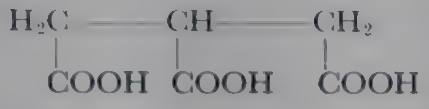
$$\cdot\mathrm{CH_2COOH} + \cdot\mathrm{CH_2COOH} \rightarrow \underset{\mathrm{COOH}}{\underset{|}{\mathrm{CH_2}}}\!\!-\!\!\!-\!\!\underset{\mathrm{COOH}}{\underset{|}{\mathrm{CH_2}}}$$

Further hydrogen abstraction from the CH_2 group of succinic acid produces a radical which can combine with a hydroxyl radical to form malic acid



$$\underset{\mathrm{COOH}}{\mathrm{CH_2}}-\underset{\mathrm{COOH}}{\mathrm{CHOH}}$$

or with a CH_2COOH radical to form tricarballylic acid



$$\underset{\mathrm{COOH}}{\mathrm{H_2C}}-\underset{\mathrm{COOH}}{\mathrm{CH}}-\underset{\mathrm{COOH}}{\mathrm{CH_2}}$$

At higher doses, repeated dehydrogenation and radical recombination reactions occur, leading to the formation of malonic and citric acids as well. In the presence of air all these products except malic acid are formed.

The irradiation of lactic acid produces principally pyruvic acid. Calcium glycolate solutions produce oxalate and formate, and oxalic acid decomposes to CO_2. In the presence of oxygen some acids form peroxy acids.

The irradiation of aerated formic acid solutions with helium ions from a cyclotron produces a variety of aldehydes and acids shown in Table 16, (Garrison 1955). These products are formed by a sequence of radical reactions analogous to those discussed previously.

TABLE 16

IRRADIATION OF AERATED FORMIC ACID SOLUTIONS[1]

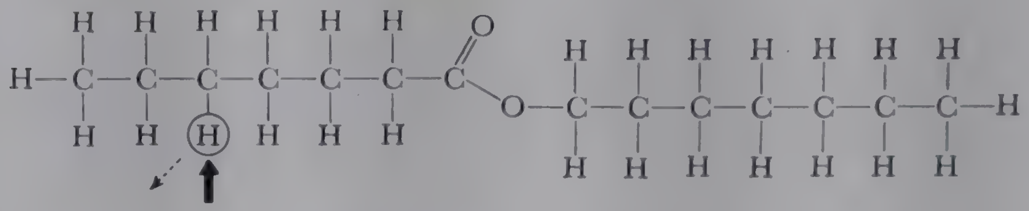
Product	Formula	*G* value
Glyoxylic Acid	$CHO(COOH)$	0.30
Mesoxalic Acid	$COH(CO)COOH$	0.20
Oxalic Acid	$(COOH)_2$	0.13
Tartronic Acid	$COOH(CHOH)COOH$	0.06
Tartaric Acid	$CO_2OH(CHOH)_2COOH$	0.006
Formaldehyde	$HCHO$	0.002
Glyoxal	$(CHO)_2$	0.12

[1] From Garrison (1955).

Some radiation induced reactions have been found which are the converse of the above processes. These include the irradiation of aqueous solutions of carbonates which produce small amounts of oxalate ion, and the irradiation of hydrogen-methane-carbon dioxide-water vapor mixtures which produce small amounts of low molecular weight compounds including one and two carbon acids (Hasselstrom and Henry 1956).

Fats and Oils

Fats and oils are esters formed by the reaction of glycerol or long-chain alcohols with long-chain carboxylic acids. The carboxylic acids may have from about ten to thirty carbon atoms, and may contain a variable number of double bonds. From the standpoint of radiation chemistry, their behavior is a combination of long chain saturated or unsaturated hydrocarbons and a carboxylate grouping. It is found that irradiation of oxygen-free fats produces primarily hydrogen gas, small amounts of methane and still smaller traces of alkanes up to butane. In this respect the behavior is very similar to long chain hydrocarbons. The hydrogen gas is produced by both a molecular and a free radical mechanism. The former mechanism leads to the formation of double bonds, or unsaturation along the chain, and the latter leads initially to positions on the chain with a free valence.



This radical can react with another radical to form a cross link between two such long oil or fat molecules (recombination) or transfer one hydrogen atom to (or from) the other radical to produce a saturated compound and a double bond (disproportionation). A third, short term possibility exists. In order for either of the above reactions to take place it is necessary that the reaction sites of two long chain molecules come into close proximity. In the liquid state of low molecular weight compounds, the molecules have sufficient mobility to accomplish this reorientation at a rapid rate. In a highly viscous oil or a solid fat, however, the mobility of the molecules is very much reduced. Hydrogen atoms can move around in the interstices of the viscous medium fairly readily but the rate of reorientation of the long chain molecules is very slow. As a consequence the active sites may persist for hours or even days (Hannan and Shepherd 1954).

The behavior under irradiation of the carboxylate part of the molecule is quite similar to that of low molecular weight acids discussed above. The principal effect of radiation is the destruction of the group with the formation of CO_2 and lesser amounts of CO.

In the presence of oxygen the radiation effects are again parallel to those discussed earlier. The first compounds formed are hydroperoxides and peroxides which subsequently lead to the production of aldehydes and ketones. In this subsequent process the long chains are generally ruptured and lower molecular weight oxidation products result. In general the materials containing double bonds (unsaturated oils) are more easily oxidized because of the particularly weak C–H bond found adjacent to the double bond.

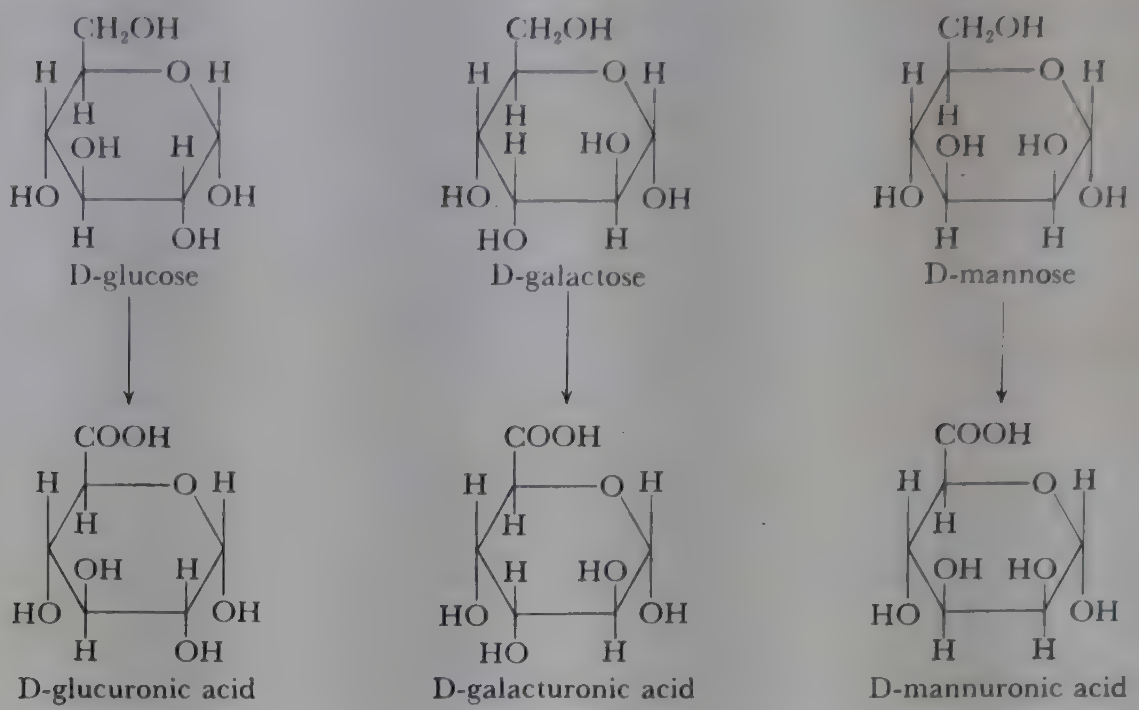
The radiation-induced oxidation of these compounds is of technological significance (Schweigert and Doty 1958). It is believed that undesirable flavors in fats and oils are due to the aldehyde and ketone formation, both in natural and in radiation-induced oxidation. Addition of antioxidants prior to irradiation reduces the formation of oxidized product, but does not always produce a corresponding reduction in undesirable flavor. The problem here is one of irradiating a highly viscous material already containing oxygen. The effectiveness of additives is not as high as desired because the diffusion rate is so small that they cannot effectively compete for reactive sites, or remove the trapped oxygen very rapidly. Further, the occurrence of trapped active sites in fats means that even the advantages of irradiation under air-free conditions may be vitiated by slow oxidation during subsequent storage. Although chemical changes are detectable after fairly small radiation doses, the flavor problem arises in the higher dose range of several hundred thousand to one million rads.

Carbohydrates (Mono and Disaccharides)

In their chemical functionality, monosaccharides are a combination of alcohol and aldehyde or ketone; they are accordingly termed aldoses and ketoses respectively. In aqueous solution it is found that an oxidation occurs which is sometimes surprisingly specific (Phillips 1954). Three six-carbon aldoses in aqueous solution, D-glucose, D-galactose and D-mannose, all produced only the acid formed from oxidation of the terminal primary hydroxyl group, see Fig. 35. The reactions are the result of oxidation by oxidizing radicals formed in the water, probably the hydroxyl radical. In contrast, a ketose in aqueous solution, D-fructose, yielded on irradiation a number of reducing sugars but no acids. This may reflect the greater resistance against oxidation of ketones as compared to aldehydes. Irradiation of the aldoses in the crystalline state produces similar changes but to a lesser extent. There are some indications of traces of formaldehyde production in some irradiated sugars, indicating carbon-carbon bond cleavage.

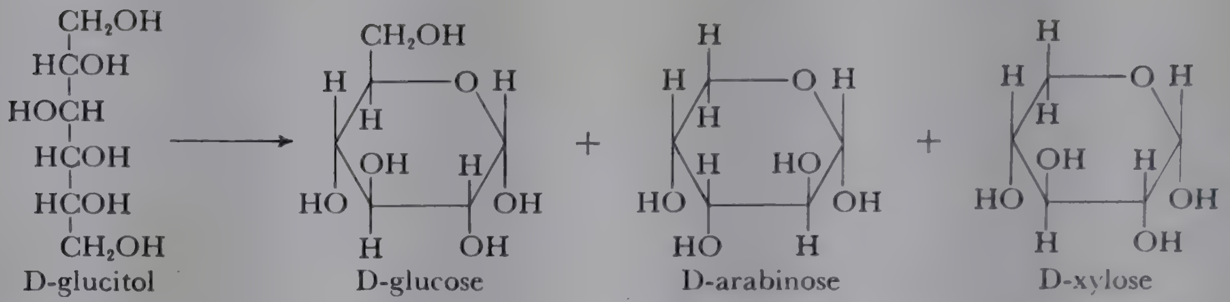
The irradiation of a number of polyhydroxy alcohols, in aqueous solution produces a variety of sugars. For example, the hexahydroxyl alcohol D-glucitol on irradiation yields both six- and five-carbon sugars, (see Fig. 36). Corresponding products, including acids, have been reported for other alcohol precursors.

In the case of aqueous solutions of disaccharides, an important radiation induced reaction is their hydrolysis to monosaccharides. Irradiation of sucrose solutions produces glucose and fructose, the monosaccharides of which sucrose is formed, as well as small amounts of other substances. Hydrolysis depends strongly on temperature, the amount increasing at


D-glucose
D-galactose
D-mannose
D-glucuronic acid
D-galacturonic acid
D-mannuronic acid

Courtesy of Phillips

FIG. 35. ACTION OF ELECTRONS ON HEXOSES


D-glucitol
D-glucose
D-arabinose
D-xylose

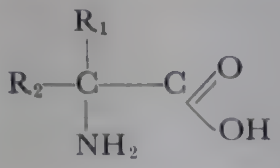
Courtesy of Wolfrom

FIG. 36. ACTION OF CATHODE RAYS ON D-GLUCITOL

lower temperature. At room temperature, in 50 per cent aqueous solutions, about one per cent hydrolysis occurs for every five million rads up to doses of one hundred million rads. The observation of radiation-induced hydrolysis is of practical significance. In the respiratory cycles of fruit and vegetable tissues it is necessary that polysaccharides be first hydrolyzed to monosaccharides before oxidation takes place. This is normally accomplished by enzymes. The irradiation of disaccharide-containing tissues produces a sudden increase in available monosaccharides and this may temporarily accelerate the respiratory rate (Burns and Desrosier 1957).

Amino Acids

The amino acids are the building blocks of the high molecular weight proteins. Hydrolysis of proteins yields some 20 different amino acids which have in common the structural features shown below. The substituents R_1 and R_2 may range in complexity from hydrogen atoms, in



which case one obtains the simplest amino acid, glycine (or amino-acetic acid), to longer chains or 5 or 6 membered ring and chain combinations. Besides carbon and hydrogen, these substituents may also contain nitrogen, oxygen or sulfur bearing groupings. The radiation effects in this class of compounds include both general effects reflecting their common amino acid structure, and more or less specific differences related to the nature of the substituent groups R_1 and R_2. It is therefore not surprising that knowledge of the radiation chemistry of these compounds is incomplete, and less so mainly for the simple representatives of the class such as glycine $(NH_2)CH_2COOH$ and alanine $(CH_3)(NH_2)CHCOOH$.

A principal result of irradiation of aqueous solutions of amino acids is the deamination of the acids to form free ammonia (Dale 1949), shown in Table 17. The mechanism is not yet clear but probably involves direct deamination resulting from attack by H· and ·OH radicals. In sulfur-containing amino acids, irradiation produces liberation of hydrogen sulfide or alternately oxidation of the S–H bonds to disulfide and finally sulfinic $-SO_2H$ and sulfonic $-SO_3H$ acid groupings. These reactions appear to proceed preferentially to deamination (Dale and Davies 1951).

TABLE 17

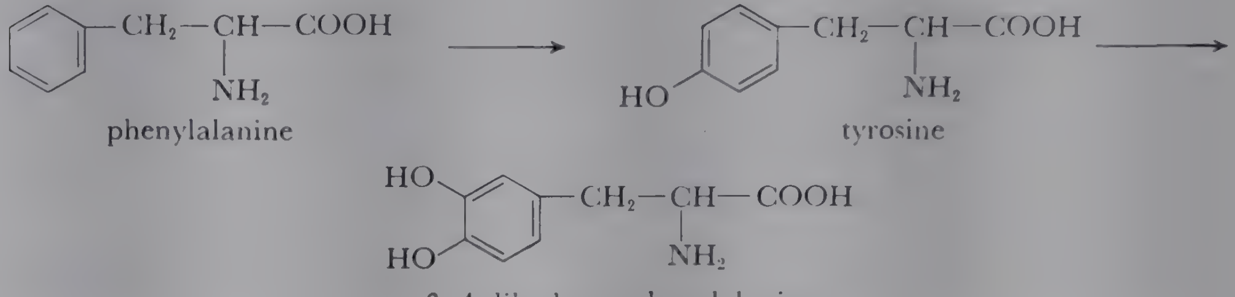
AMMONIA YIELD FROM VARIOUS AMINO ACIDS AND OTHER NITROGEN CONTAINING SUBSTANCES (0.13*M* solutions, 150,000 rads)[1]

Compound	Micrograms ammonia per ml
glycine	8.1
glycine plus HCl	5
glycine plus NaOH	8
Alanine	7.2
B-alanine	4.3
Glycylglycine	12.5
Glycylglycine plus HCl	3.8
Leucylglycine	6.6
Diglycylglycine	7
Proline	0
Urea	0.2
Thiourea	1.4

[1] From Dale (1949).

The deamination process produces a radical formed by removal of the NH_2 group from the α carbon atom, and the subsequent reaction of the radical in irradiated aqueous solution produces a variety of oxidation products. These reactions are in some respects analogous to those discussed above in connection with alcohols and acids. For example glycine $(NH_2)CH_2COOH$, produces formaldehyde, $HCHO$, alanine, $(CH_3)(NH_2)CHCOOH$, produces acetaldehyde CH_3CHO, and serine $(CH_2OH)(NH_2)CHCOOH$ produces glycolaldehyde $(CHO)(CH_2OH)$ and glyoxal $(CHO)(CHO)$.

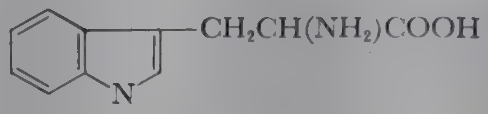
In an aqueous solution of the ring-containing amino acid phenylalanine there occurs a hydroxylation of the ring, presumably by hydrogen abstraction followed by addition of a hydroxyl radical (Proctor and Bhatia 1952). This leads to another amino acid tyrosine which in turn can undergo further hydroxylation.


phenylalanine

tyrosine

3, 4-dihydroxy phenylalanine

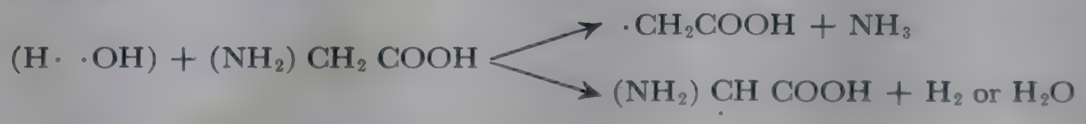
This reaction has its close parallel in hydrocarbon radiation chemistry in the production of phenol C_6H_5OH from benzene C_6H_6 in aqueous solution. Analogous reactions are found in tryptophan, which has a fused ring substituent.


tryptophan

Irradiation produces hydroxylation of the six membered ring among other things.

The best understood system is that of aqueous glycine solutions. The mechanism proposed by Garrison and Weeks (1956) for this system is as follows:

Hydrogen and hydroxyl radicals act on glycine in two ways: removal of the amino group or removal of a hydrogen from the amino-bearing carbon



$$(\mathrm{H}\cdot\ \ \cdot\mathrm{OH}) + (\mathrm{NH_2})\ \mathrm{CH_2\ COOH} \begin{cases} \rightarrow \cdot\mathrm{CH_2COOH} + \mathrm{NH_3} \\ \rightarrow (\mathrm{NH_2})\ \underset{\cdot}{\mathrm{C}}\mathrm{H\ COOH} + \mathrm{H_2} \text{ or } \mathrm{H_2O} \end{cases}$$

The nitrogen containing radical may be formed with the alternate structure $(\cdot NH)CH_2COOH$. Recombination of the radicals with one another (or ones of the same type) results in the formation of succinic acid $(\mathrm{CH_2COOH})_2$, amino succinic acid $(\mathrm{CH_2COOH})(\mathrm{CHNH_2COOH})$ and diamino succinic acid $(\mathrm{CHNH_2COOH})_2$. These products have been identified under high dose rate conditions. At lower dose rates the carboxymethyl radical $\cdot\mathrm{CH_2COOH}$ abstracts a hydrogen from glycine to form acetic acid and another starting radical. The nitrogen bearing radical can recombine, as already discussed, or undergo the following reactions

$$\mathrm{H_2O_2 + NH_2CHCOOH} \xrightarrow[\text{oxidation}]{} \mathrm{H_2O} + \cdot\mathrm{OH} + \underset{\substack{\text{iminoacetic acid}\\ \text{(unstable)}}}{\mathrm{NHCHCOOH}}$$

$$\mathrm{NHCHCOOH + H_2O} \xrightarrow[\text{hydrolysis}]{} \mathrm{NH_3} + \underset{\text{glyoxylic acid}}{\mathrm{CHO\ COOH}}$$

or

$$\mathrm{NHCHCOOH + H_2O} \xrightarrow[\text{hydrolysis}]{} \mathrm{NH_3 + HCHO + CO_2}$$

Finally, the nitrogen-bearing radical can undergo disproportionation to form glycine and unstable imino acetic acid which is hydrolyzed as above

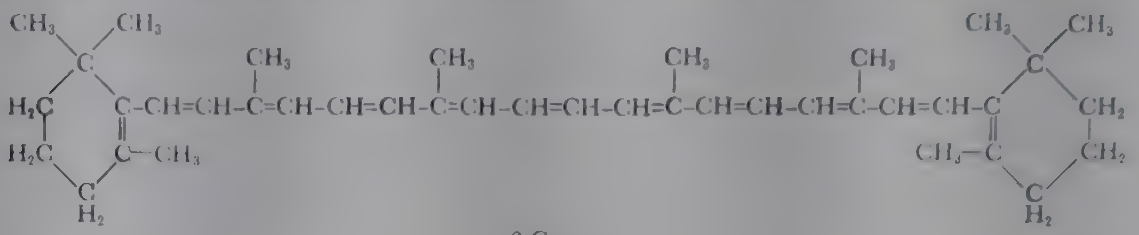
$$2\ \mathrm{NH_2\underset{\cdot}{C}HCOOH} \rightarrow \mathrm{NH_2CH_2COOH + NHCHCOOH}$$

In summary, glycine radiolysis forms ammonia, acetic acid, glyoxylic acid, succinic acid, amino succinic acid, diamino succinic acid, formaldehyde, hydrogen and carbon dioxide. All these are accounted for by the above mechanism. Further support for the mechanism is found in the radiolysis of alanine. This substance is derived from glycine by substituting a methyl group for a hydrogen atom on the ammonia bearing carbon. The products found on radiolysis differ accordingly in structure, e.g. acetaldehyde instead of formaldehyde, but their relative yields are surprisingly parallel. Some further fine points in the problem not covered by the above mechanism are discussed in the literature (Garrison and Weeks 1956). The radiation behavior of polypeptides and proteins will be discussed in the next chapter.

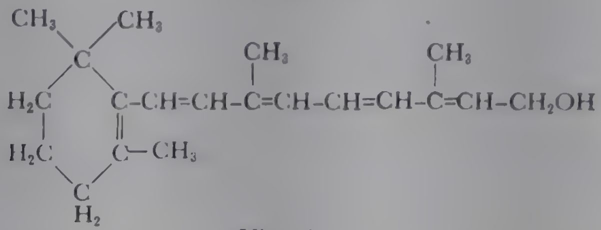
Vitamins

The exposure of vitamins to ionizing radiations can be expected to have subtle but significant effects. The specific molecular changes which occur vary from compound to compound and the conditions and environment of irradiation. Both direct and indirect effects are found. The vitamins are classified on the basis of their solubility in fat or water, and will be discussed in this order.

Fat Soluble Vitamins.—Several carotene pigments are found in the plant kingdom which are converted to vitamin A in the animal body. Beta-carotene yields two molecules of the vitamin A while alpha and gamma carotenes yield one. The conversion of carotene involves splitting of the hydrocarbon chain at its central double bond, to form vitamin A aldehyde, which is then reduced to its alcohol. Carotenes are termed provitamin A.



β-Carotene



Vitamin A

Animal fats contain the vitamin itself, either as a free alcohol or as the ester of higher fatty acids, while plant fats contain carotene. Milk fat is an exception in that it contains both the vitamin and carotene. Fish liver oils contain vitamin A in ester form combined with aliphatic acids.

TABLE 18

STABILITY OF CAROTENOIDS TO RADIATION[1]

Source	Carotenoid	Substrate of Carotenoid	Per Cent Destruction (2×10^6 rads)
Shrimp	Astaxanthin	Bound to protein	25
Salmon	Astaxanthin	Highly unsaturated lipids	98
Chicken	Lutein dipalmitate	Lipid with some unsaturation	55
Beef	Carotene	Lipid with some unsaturation	65

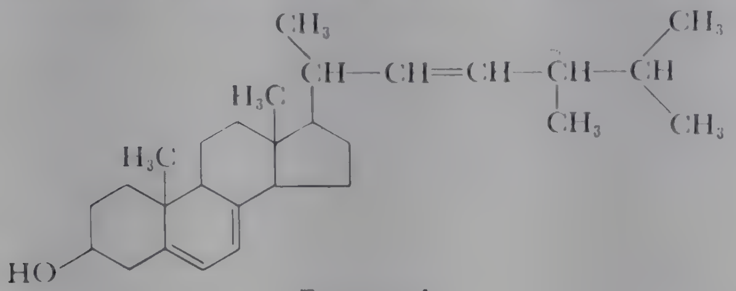
[1] Adapted from Sheffner and Spector (1957).

The sensitivity of vitamin A to irradiation is influenced by the media in which it is exposed as shown in Table 18. The difference in sensitivity is not explainable in terms of moisture present, indicating that a substantial destruction occurs due to direct hit mechanisms. The greater sensitivity of Vitamin A in butter is perhaps related to its association with unsaturated fatty acids. In margarine, however, the vitamin A esters used are more stable to radiation effects. The importance of the irradiation medium is also shown in the greater destruction of beta carotene in petroleum ether than in the tomato.

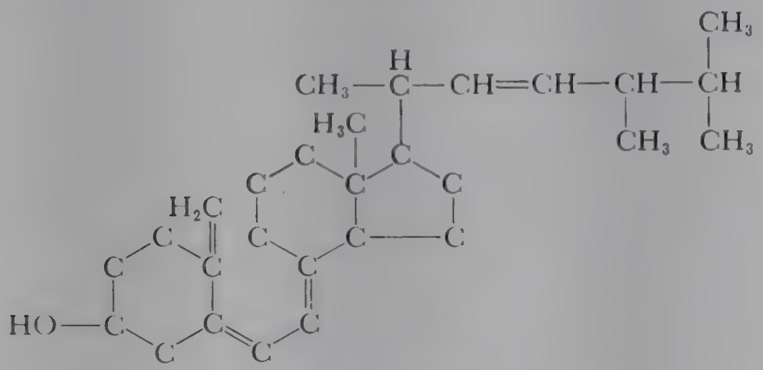
It has been shown that the substrates with which carotenes are associated are of great importance in altering radiation sensitivity. In shrimp, the carotenoid astaxanthin is attached to protein, and is rather stable to radiation effects. The astaxanthin in salmon on the other hand is not so tied, but is associated with highly unsaturated lipids. See Table 18.

The destruction of vitamin A in human blood is also small, indicating the presence of a protective mechanism.

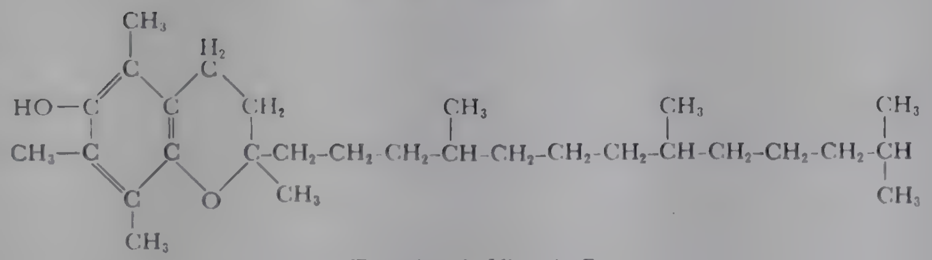
Vitamin D is obtained by the ultra-violet treatment of ergosterol, yet its irradiation with gamma photons finds but traces of vitamin D formed. In fact, exposures to gamma photons results in destruction of the vitamin D present. Irradiation of feed for chicks at 3 million rads results in some destruction of vitamin D, as shown by means of biological assay.



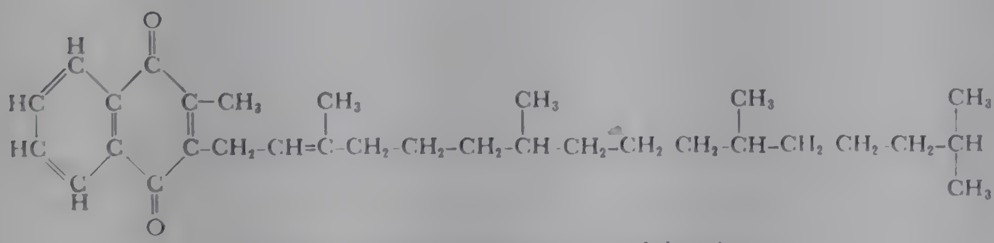
Ergosterol



Calciferol (Vitamin D_2)



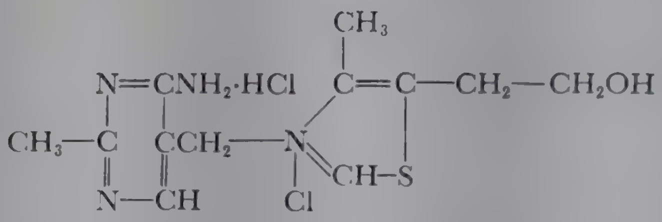
α-Tocopherol (Vitamin E)



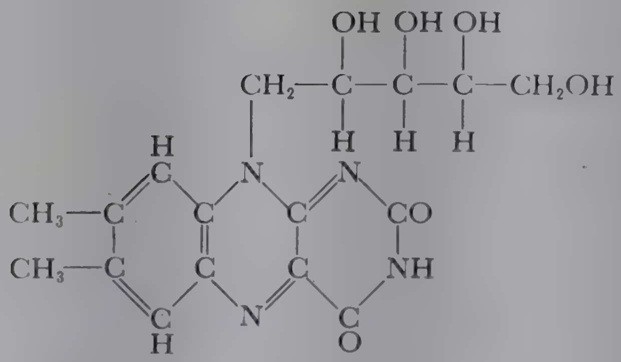
Vitamin K_1 (2-methyl-3-phytyl-1, 4-naphthoquinone)

Vitamin E and vitamin K are both sensitive to ionizing radiations, the latter being nearly completely destroyed with doses over 3 million rads. The vitamin K activity of fresh spinach on the other hand was reported by Sheffner and Spector (1957) to be radiation resistant. Large losses in vitamin E also occur. The addition of these to the diet of growing animals remedies the deficiency, indicating the specificity of the reaction, which is characteristic of the vitamins as a group. Vitamin E and K irradiated in the absence of oxygen are more stable than in its presence.

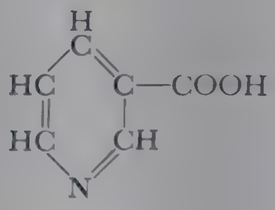
Water Soluble Vitamins.—The water soluble vitamins embrace a large number of essential nutrients for man. Originally vitamin B was named for its ability to prevent beriberi in man. Subsequent studies uncovered a series of twelve and more components, hence the name B complex. The chemical entities which constitute the class are not related one to another either chemically or physiologically, but as a class they are related in that they are part of enzyme systems which play a vital role in metabolism.



Thiamine Hydrochloride
(Vitamin B_1)



d-Riboflavin



Niacin
(Nicotinic Acid)

Vitamins in the B complex are generally radiation sensitive, but vary in their destruction by exposures to ionizing radiations. Thiamin is notably sensitive to the effects of heating generally, and destruction by radiation can be demonstrated to be dose dependent. The retention of thiamin in pork and beef with increasing dose applications is shown in Table 19. While riboflavin is easily destroyed by visible light, exposures to gamma photons finds the vitamin more resistant than thiamin. The

Table 19

RADIATION EFFECTS ON THIAMIN, RIBOFLAVIN, AND NIACIN CONTENT OF BEEF AND PORK[1]

Dose Megarads	Beef Thiamin (mg./100 gm.)	Beef Riboflavin (mg./100 gm.)	Beef Niacin (mg./100 gm.)	Pork Thiamin (mg./100 gm.)	Pork Riboflavin (mg./100 gm.)	Pork Niacin (mg./100 gm.)
0	0.079	0.15	4.5	0.96	0.27	4.6
0.3	.085	0.15	4.3	0.99	0.31	4.3
1	.065	0.15	4.3	0.97	0.27	4.3
3	.015	0.13	3.4	0.12	0.19	3.6
10	.006	0.13	3.3	0.04	0.20	3.6

[1] From Sheffner and Spector (1957).

destruction of riboflavin at a given dose increases with increasing concentration, but very high concentrations are more stable. Water soluble niacin is nearly as radiation resistant as riboflavin (Table 19).

The radiation sensitivity of the vitamins in the B complex is influenced by the conditions of irradiation. Riboflavin is rather stable in the presence of ascorbic acid, and in milk substrates is more than ten times as stable as in pure water solutions (Kung *et al.* 1953).

Niacin retention during irradiation is greater than thiamin in pork and beef, as shown in Table 19. The extent of destruction varies with concentration, high levels (100 mg/ml) being rather radiation resistant in water solutions, low concentrations (20 mg/ml) being inactivated at low dosages.

Using carbon-14 tagged niacin, Goldblith *et al.* (1949) have shown that decarboxylation occurs first, followed by splitting of the pyridine ring. King and Becker (1955) found upon the irradiation of a nicotinamide that shifts in absorption spectra occur.

Vitamin C, ascorbic acid, is required to prevent scurvy and is found widely in fresh fruits and vegetables, succulent tissues usually being excellent sources. Freshly killed flesh soon loses antiscorbutic value.

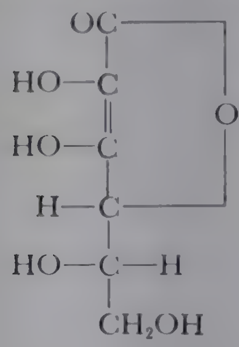
Table 20

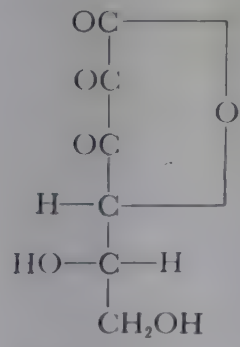
RADIATION EFFECTS ON ASCORBIC ACID IN PLANT TISSUES EXPOSED TO 2×10^6 RADS UNDER SEVERAL CONDITIONS[1]

Plant Tissue	Treatment	Ascorbic Acid mg./100 gm.	Ascorbic Acid Per cent retention
Broccoli	Fresh control	18	100
	Irradiated fresh	2.5	14
	Irradiated frozen	6.0	33
	Irradiated in vacuum	1.8	10
Spinach	Fresh control	6.2	100
	Irradiated fresh	2.2	36
	Irradiated frozen	0.4	7
	Irradiated in vacuum	1.2	20

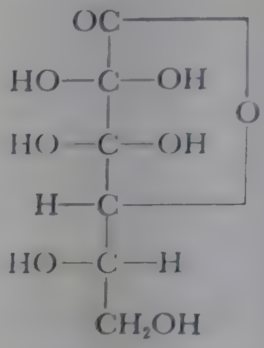
[1] From Sheffner and Spector (1957).

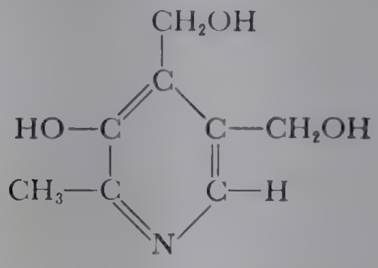
The irradiation of ascorbic acid in aqueous solution finds it radiation sensitive. In dilute concentrations, 4–5 μg/ml., nearly complete destruction is found at 10,000 rads. In more concentrated solutions (100 μg/ml.) 100,000 rads causes the destruction of slightly more than 50 per cent. Freezing protects the vitamin to some extent (Table 20). In native materials such as citrus juice, the vitamin is more radiation resistant than in water alone. Yet, it is commonly found that vitamin C is one of the first to be destroyed upon irradiation. This has led to the use of this vitamin, of known antioxidant properties, as an additive to protect irradiated substrates.

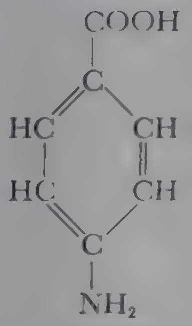

Vitamin C—Ascorbic Acid

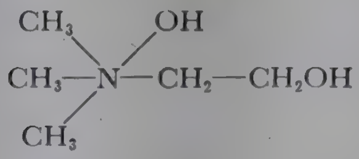

Ketone Formula

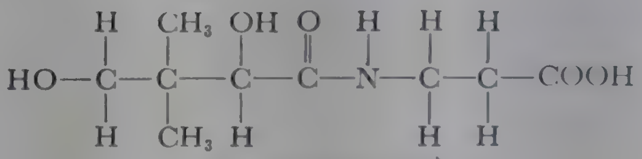
Dehydroascorbic Acid


Hydrate Formula


(Pyridoxine)

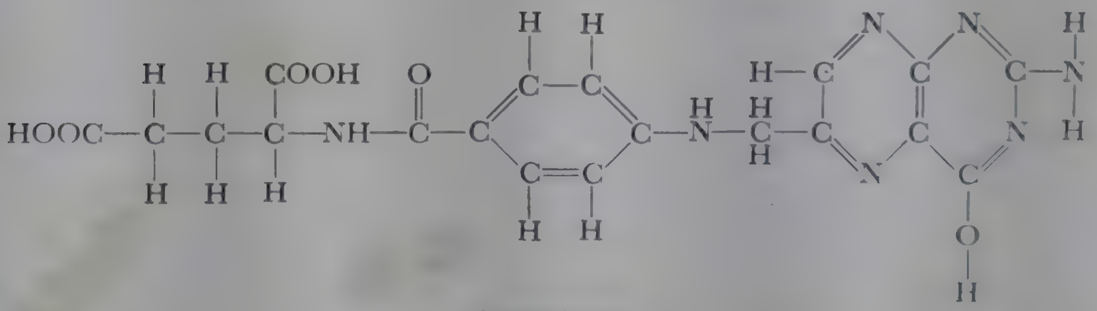

p-amino-benzoic acid


Choline



$$HO-\overset{H}{\underset{H}{C}}-\overset{CH_3}{\underset{CH_3}{C}}-\overset{OH}{\underset{H}{C}}-\overset{O}{\overset{\|}{C}}-\overset{H}{N}-\overset{H}{\underset{H}{C}}-\overset{H}{\underset{H}{C}}-COOH$$

Pantothenic Acid


Folic Acid

N-4 [(2-amino-4-hydroxy-6-pteridy) methyl amino] benzoyl glutamic acid

Exposures of ascorbic acid to 6 and 12 million rads produces dehydroascorbic acid, diketogulonic acid and some carbon dioxide (Scheffner and Spector 1957). Dehydroascorbic acid retains antiscorbutic activity, well known to be reversible biologically by animals into ascorbic acid. The usefulness of ascorbic acid in protecting other nutrients has been shown above. Its value in protecting protein in solution from radiation damage is less effective, although protection is afforded to some extent by suitable additions, as will be shown later.

Vitamin B_6 (pyridoxine) has the same order of radiation sensitivity as does riboflavin. The para amino benzoic acid molecule is not altered easily with doses of less than a million rads, after which decarboxylation and deamination occur. Pantothenic acid is destroyed only to the extent of some 10 per cent after exposures of animal flesh to 6×10^6 rads, and the potency of folic acid is not diminished when so treated. Choline is also relatively stable.

TABLE 21

COMPARATIVE VITAMIN RETENTION IN CANNING AND RADIATION PRESERVATION OF FOOD[1]

	Per cent Retention in Food	
Vitamin	Canning	Radiation Stabilization
Thiamin	35	35
Riboflavin	80	80
Pyridoxine	70	75
Niacin	75	75
Folic acid	70	90
Vitamin A	80	75
Vitamin E	90	75
Vitamin K	90	15

[1] Adapted from Progress Report on Atomic Energy Research 84th Congress of the United States.

The comparative vitamin retention in food preserved by irradiation as compared with food preserved by canning is seen in the following Table 21. Considering the youthfulness of the art of irradiation, it may be anticipated that significant improvements in nutrient protection will be made in the future.

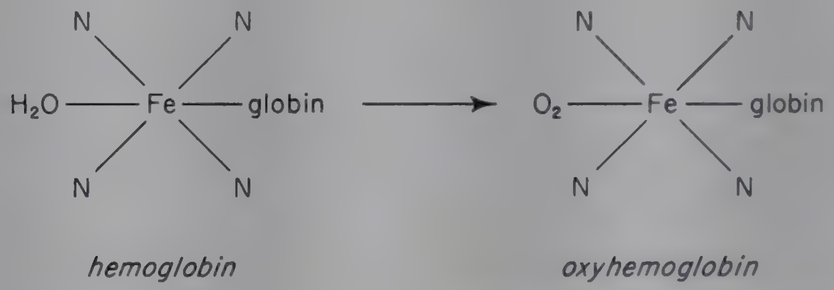
Pigments

Considering the rather delicate chemical nature of pigments, it is to be anticipated that their exposure to ionizing radiation will have noticeable effects. The pigments in red meats have received substantial attention, due mainly to the pronounced effects radiation produces. The irradiation of plant tissues containing pigments also induces recognizable chemical changes in several instances.

Pigments of Animal Flesh.—Hemoglobin is composed of a protein (Globin) and an iron-containing component called heme. Myoglobin is a component of muscles resembling hemoglobin in ability to combine with oxygen. The exposure of such chemical entities to ionizing radiation may be anticipated to alter chemical and molecular configurations, hence have effects on their ability to impart color.

The pigments of red meat are principally hemoglobin and myoglobin. The content of the former is dependent upon the amount of residual blood left in the tissue, while myoglobin is an integral component of muscle tissue. These two pigments are closely related chemically, and reactions differ in degree rather than kind.

In fresh beef hemoglobin exists in two forms depending on the presence or absence of oxygen. The oxygenation of hemoglobin converts it to oxyhemoglobin, shown schematically in Figure 37. The pigment myoglobin also occurs as a compound itself, or in the presence of oxygen, as oxymyoglobin. Oxymyoglobin is present in surfaces of meat exposed to the air, while internal tissues have reduced myoglobin present. Oxymyoglobin has a bright red color and gives beef its characteristic bright color; myoglobin is a dark purple and is the color seen in freshly cut tissue. Normally after being exposed to the air, myoglobin will become oxygenated and turn a bright red.



Courtesy of Schweigert and Doty

FIG. 37. SCHEMATIC DIAGRAM OF RADIATION EFFECT ON HEMOGLOBIN

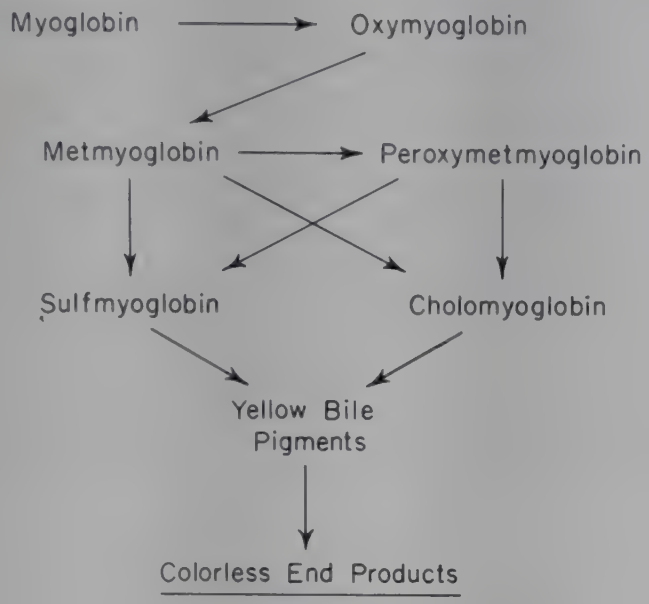
The color of irradiated beef is due primarily to two pigments, one red and the other green, products of free radical oxidation of myoglobin. Usually the red color predominates, and only under aerobic conditions does sufficient green pigment appear to be noted, if seen. There are also yellow pigments which are extractable from irradiated beef, thought to be breakdown products of heme. If sufficiently irradiated, upwards of 100 megarads, all pigments can be destroyed. At 6 megarads some 20 per cent of the total pigmentation may be lost.

Studies on myoglobin, mainly at the American Meat Institute, reveal that gross changes in the red color of meats occur when oxygen is present in both heme and globin moieties. On an overall basis, the following sequence of events is believed to occur (Schweigert and Doty 1958) (Fig. 38).

The nature and quantity of products formed are dependent on the pH of the reaction, presence of reducing agents or sulfhydryl groups, the concentration of myoglobin, the dose applied, and other factors.

The maximum green pigment formed from irradiated metmyoglobin solutions occurs near a pH of 5.2.

Gamma photons can reverse the reaction, oxyhemoglobin being obtained from methemoglobin, which may be an explanation for the bright red color of irradiated fresh meats.



Courtesy of Schweigert and Doty

FIG. 38. RADIATION EFFECTS OF PIGMENTS OF RED MEAT

In raw meat, irradiated myoglobin is oxidized to oxymyoglobin even in the absence of oxygen and a bright red color is obtained. With oxygen present, a brown color is obtained due to metmyoglobin. In cooked irradiated meat, the main reaction is a reduction of hemichrome to hemochrome resulting in a bright red color, not ordinarily seen in a cooked piece of beef. However, upon exposure to air the meat pigments quickly convert and the gray-brown normal appearance of cooked meat prevails.

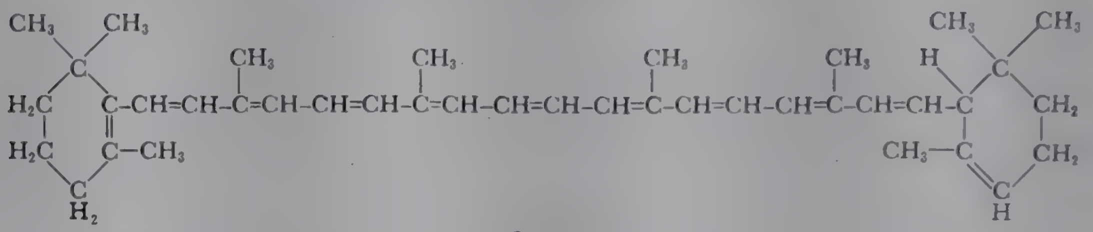
Chicken flesh remains pink after being irradiated and cooked, with the same explanation. Upon exposure to air, irradiated chicken flesh too loses its pinkness. Pork flesh behaves similarly and is of more significance, because of the widely held notion that pork cooked so that it is still pink internally is inedible (due to the fear of ingesting live trichina). However, Trichinella is destroyed by the irradiation process.

Pigments of Plant Tissues.—Chlorophyll in plants is associated with carotenes and xanthophylls (dihydroxyl derivatives of carotenes). The irradiation of peas cause losses in chlorophyll, and the presence of pheophytin. The amount of pheophytin found in this instance is correlated with the decrease in chlorophyll pigments.

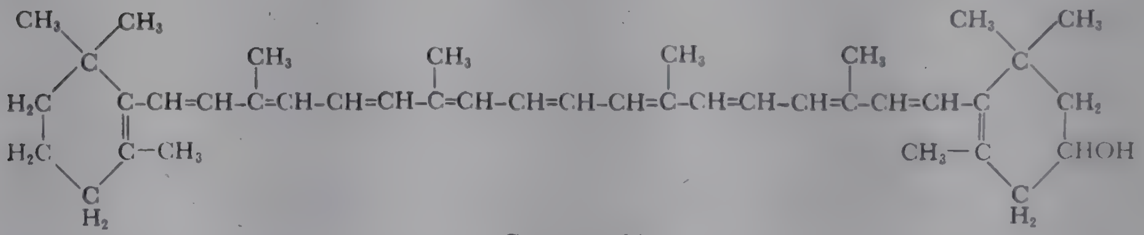
Heating peas prior to irradiation increases the development of pheophytin, high temperatures causing substantial color destruction alone. At 2 × 10^6 rads, there is a loss of about 10 per cent of the chlorophyll present in green bean pods (Morgan and Siu 1957). Pheophytin is not formed as the degradation product.

Heating chlorophyll-containing plant tissues results in the loss of some pigment magnesium, converting chlorophyll to pheophytin which corresponds to a color change from grass green to olive green. Upon irradiation of green plants, chlorophyll is bleached and the mechanism of reaction induced by irradiation requires further investigation.

The carotenoids have characteristically several unsaturated carbon bonds, which are susceptible to oxidation, yet the pigments are relatively stable in natural plant tissue substrates, in comparison with their radiation sensitivity in dilute extracts or pure solutions.



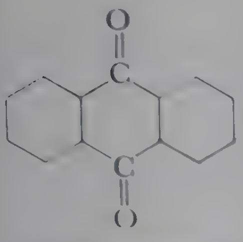
α-Carotene

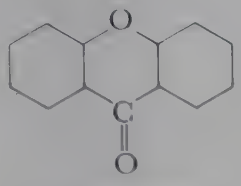


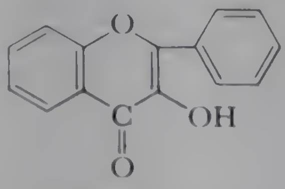
Cryptoxanthin

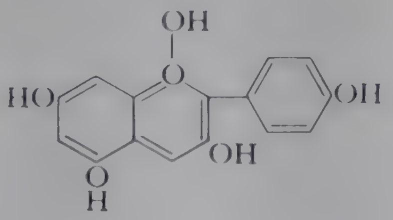
Lycopene, the red pigment of the tomato, can be chemically altered by irradiation. The development of the normal red pigmentation is inhibited by radiation treatments of two million rads. Fruits irradiated only in a band around their equator with cathode rays show a normal color development over the unexposed blossom and stem ends, with inhibition of normal pigmentation in the exposed areas (Burns and Desrosier 1957).

The water soluble red, blue, and violet pigments of fruits and plants are generally anthocyanins. Flavones are closely related yellow compounds. Anthocyanins are pH indicators, being red in acid and blue in alkaline solutions. On oxidation, color changes occur from violet-red to brick-red to brown. Complete bleaching is obtained by exposing red anthocyanins to sulfur dioxide, and when it is removed from solution, the color returns. The anthocyanins are generally bleached by irradiation, however, it is not uncommon to find the pigments recover their normal color following a period of storage. With fruits given dosages less than a million rads, the initial bleaching is overcome and regeneration or resynthesis of pigments occurs.


Anthraquinone (Red derivatives)


Xanthone (Yellow derivatives)


Flavonol (Yellow derivatives)


Anthocyanin nucleus (Red, blue, and violet derivatives)

Strawberries recover from the irradiation at levels of 200,000 rads, then continue to increase in pigment content in normal fashion. At four million rads, there are irreversible pigment losses.

The irradiation of pigments in natural fruit substrates which are frozen during irradiation protects the pigments, less color change being noted than when irradiation occurs in unfrozen tissues (Morgan and Siu 1957).

BIBLIOGRAPHY

Astrack, A., Sorbye, O., Brasch, A., and Huber, W. 1952. Effects of high intensity electron bursts upon various vegetable and fish oils. Food Research *17*, 571–575.

Bakh, N. A. 1959. Role of Oxygen and Peroxides in Radiation Chemistry. Symposium B. Radiation Research Supplement No. 1, 149–215.

Bakh, N. A., Medvedovsky, V. I., and Saraeva, V. V. 1958. Radiolysis and radiation induced oxidation of organic substances. Geneva Conference *29*, 128–134.

Bell, E. R., Raley, J. H., Rust, F. F., Seubold, F. H., Jr., and Vaughan, W. E. 1953. Discussions Faraday. Soc. 10, 246.

Burns, E. E., and Desrosier, N. W. 1957. Maturation changes in tomato fruits induced by ionizing radiation. Food Technol. *11*, 313–317.

Burr, J. G. 1957. The radiolysis of deuteriated and tritiated acetic acids. J. Phys. Chem. *61*, 1481–1483.

Burton, M., and Lipsky, S. 1957. Mechanisms of protection in radiolysis of organic systems. J. Phys. Chem. *61*, 1461–1466.

Burton, M., Chang, J., Lipsky, S., and Reddy, M. P. 1957. Radiolysis of mixtures: cyclohexane-benzene-iodine. J. Chem. Phys. *26*, 1337–1338.

Chevallier, A., and Burg, C. 1955. Action of ionizing radiation on lipids. Radiobiology Symposium, Liége, 1, Belgium.

Chang, P. C., Yang, N. C., and Wagner, C. D. 1959. Direct dimerization of terminal olefins by ionizing radiation. J. Am. Chem. Soc. *81*, 2060–2064.

Collison, E., and Swallow, A. J. 1956. The radiation chemistry of organic substances. Chem. Rev. *56*, 471–568.

Dale, W. M., and Davies, J. W. 1951. The liberation of hydrogen sulfide by X-radiation from cysteine and glutathione. Biochem. J. *48*, 129–134.

Dale, W. M., Davies, J. V., and Gilbert, C. W. 1949. The kinetics and specificities of deamination of nitrogenous compounds by x-radiation. Biochem. J. *45*, 93–96.

Dewhurst, H. A. 1957. Radiation chemistry of organic compounds I. n-alkane liquids. J. Phys. Chem. *61*, 1466–1471.

Dewhurst, H. A. 1959. Radiation chemistry of organic compounds. IV. Cyclohexane. J. Phys. Chem. *63*, 813–818.

Dorfman, L. M., and Shipko, F. J. 1955. The Beta-particle radiolysis of acetylene. J. Am. Chem. Soc. *77*, 4723–4728.

Drake, M. P., and Giffee, J. W. 1957. Action of ionizing radiation on amino acids, proteins and nucleoproteins. Chap. 17, Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

Elliott, A. M., Brownell, L. E., and Gross, J. A. 1954. Use of Tetrahymena to evaluate the effects of X-radiation on essential nutrilites. J. Protozool. *1*, 193–197.

Garrison, W. M. 1955. Irradiation of formic acid. UCRL-3013, 48, July–Sept.

Garrison, W. M., Bennett, W., Cole, S., Haymond, H. R., and Weeks, B. M. 1955. Indirect and direct action of heavy particle radiation on acetic acid in aqueous solution. J. Am. Chem. Soc. *77*, 2720–2727.

Garrison, W. M., and Weeks, B. M. 1956. Mechanisms in the radiolysis of of aqueous glycine solutions. J. Chem. Phys. *24*, 616–618.

Goldblith, S. A., Proctor, B. E., Hogness, J. R., and Langham, W. H. 1949. The effect of cathode rays produced at 3000 kilovolts on niacin tagged with C^{14}. J. Biol. Chem. *179*, 1163–1167.

Hamill, W. H. 1959. Ion-molecule reactions. J. Chem. Educ. *36*, 346–349.

Hannan, R. S., and Shepherd, H. J. 1954. Some after effects in fats irradiated with high energy electron and X-rays. Brit. J. Radiol. *27*, 36–42.

Hasselstrom, T. H., and Henry, M. C. 1956. New synthesis of oxalic acid. Science *123*, 1038–1040.

Holman, R. T. 1954. Autoxidation of fats and related substances: Holman, R. T.,Lundberg, W. O., and Malkin, T., Eds. Progress in the Chemistry of Fats and Other Lipids. Academic Press, New York.

Khenokh, M. A. 1955. Action of X-radiation of cobalt-60 on carbohydrates. Doklady Akad. Nauk, U.S.S.R. *104*, 746–750.

King, C. G., and Becker, R. R. 1955. Radiation effects on vitamins. Proj. Report U.S. Army Surgeon General. Con. No. DA 49-007-MD-550, Oct. 15, 1954–Jan. 15, 1955.

Kung, H., Gaden, E. L., Jr., and King, C. G. 1953. Vitamins and enzymes in milk: effect of X-irradiation on activity. J. Agr. Food. Chem. *1* 142–146.

Lind, S. C. 1928. The Chemical Effects of Alpha Particles and Electrons. Chemical Catalog Co., New York (New edition in preparation.)

LIND, S. C., and BARDWELL, D. C. 1925. The chemical action of gaseous ions produced by alpha particles: reactions of the oxides of carbon. Science *67*, 2675–2697.

LIND, S. C., and BARDWELL, D. C. 1926. The chemical action of gaseous ions produced by alpha particles. J. Am. Chem. Soc. *48*, 1575–1583.

LIND, S. C., and RUDOLPH, P. S. 1957. Retardation of a radiochemical reaction by charge transfer in a bimolecular mixture of gases. J. Chem. Phys. *26*, 1768–1770.

LONG, L., and LIROT, G. J. 1957. Action of ionizing radiations on carbohydrates and polysaccharides, Chap. 16, Radiation Preservation of Foods, U. S. Govt. Printing Office, Washington, D. C.

MANION, J. P., and BURTON, M. 1952. Radiolysis of hydrocarbon mixtures. J. Phys. Chem. *56*, 560–568.

MCCAULEY, C. E., and SCHULER, R. H. 1957. Radical production in the radiolysis of liquid butane. J. Am. Chem. Soc. *79*, 4008–4011.

MCDONNELL, W. R., and NEWTON, A. S. 1954. The radiation chemistry of the aliphatic alcohols. J. Am. Chem. Soc. *76*, 4651–4658.

MITCHELL, J. H. 1957. Action of ionizing radiation on fats, oils and related compounds, Chap. 15, Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

MORGAN, B. H., and SIU, R. G. H. 1957. Action of ionizing radiation on individual foods, Chap. 26, Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

NEWTON, A. S. 1957. A survey of the radiation chemistry of some aliphatic ethers. J. Phys. Chem. *61*, 1485–1490.

NEWTON, A. S. 1957. Some aspects of the radiolysis of isopropyl ether. J. Phys. Chem. *61*, 1490–1496.

PHILLIPS, G. O. 1954. Action of ionizing radiation on aqueous solutions of carbohydrates. Nature *173*, 1044–1046.

POLAK, L. S., TOPCHIEV, A. V., and CHERNYAK, N. Y. 1958. Radiolysis of alkanes. Second Geneva Conference *29*, Chemical Effects of Radiation. 135–139.

PRÉVOT-BERNAS, A., CHAPIRO, A., COUSIN, C., LANDLER, Y., and MAGAT, M. 1952. Radiolysis of some organic liquids. Discussions Faraday Soc. *12*, 98–109.

PROCTOR, B. E., and BHATIA, B. S. 1952. Effect of high voltage cathode rays on aqueous solutions of tryptophan, tyrosine, phenylalanine and cysteine. Biochem. J. *51*, 535–540.

PROCTOR, B. E., and GOLDBLITH, S. A. 1949. Effect of soft X-rays on vitamins (niacin, riboflavin and ascorbic acid). Nucleonics *5*, No. 3, 56–62.

RAMSBOTTOM, J. M., GOESER, P. A., and SCHULTZ, H. W. 1951. How light discolors meat. Food Ind. *23*, 120–122.

SCHWEIGERT, B. S., and DOTY, D. M. 1958. Chemistry of color, flavor, and odor changes in irradiated meats. Research Report S-507, U. S. Quartermaster Food and Container Inst., Chicago, Ill.

SCHULER, R. H. 1958. Scavenger methods for free radical detection in hydrocarbon radiolysis. J. Phys. Chem. *62*, 37–42.

SEMENOV, N. 1958. Some Problems of Chemical Kinetics and Reactivity. Pergamon Press, New York.

SKRABA, W. J., BURR, J. G., and HESS, D. N. 1953. The decomposition of methanol-C^{14} under the influence of its own radiation. J. Chem. Phys. *21*. 1296–1298.

SHEFFNER, A. L., and SPECTOR, H. 1957. Action of ionizing radiations on vitamins, steroids, hormones and other physiologically active compounds. Chap. 18, Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

STRONG, J. B., and BURR, J. G. 1959. The radiolysis of organic solutions I. Acetone as a trap for hydrogen atoms. J. Am. Chem. Soc. *81*, 775–779.

WALLING, C. 1957. Free Radicals in Solution. John Wiley and Sons, Inc., New York.

WILLIAMS, R. R., JR., and HAMILL, W. H. 1954. Chemical effects of electron capture by solutes in hydrocarbons during gamma irradiation. Radiation Research *1*, 158–164.

CHAPTER 10

Effects of Radiation on Biological Polymers

The biological polymers may be divided into four groups: carbohydrates, proteins, enzymes, and nucleic acids. Because of the complexity of these materials, the measures of the effects of radiation are frequently incomplete from a molecular standpoint. Some of the effects consist of clearly demonstrable changes in behavior which may be traced back to the chemistry of the starting materials, while other effects such as changes in enzyme and genetic activity still lack a clear connection between function and the chemical nature of the substances (enzymes and nucleoproteins). However, it has been clearly established that the above four classes exhibit an extremely wide variation in sensitivity towards high energy radiation.

All high polymers, natural and artificial, consist of molecules having a variety of molecular weights. Thus, one must regard the molecular weight of a polymer as some sort of average.

In contrast to monomeric molecules, the determination of molecular weights of polymers is a rather difficult task. A great variety of methods are available which derive a measure of an averaged molecular weight from one or another physico-chemical property of the molecule, and, reflecting our incomplete understanding of these molecular properties, give answers which are not in exact accord. The answers are, however, frequently quite close. The properties for measuring molecular weight include:

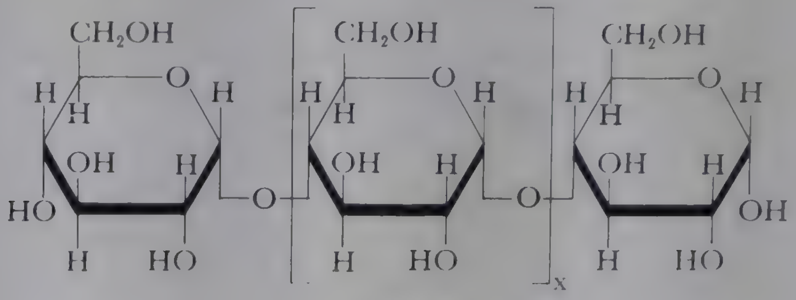
a. Viscosity in solution.
b. Osmotic pressure.
c. Light scattering.
d. Presence of a unique functional group on a polymer molecule, such as the end group.
e. Sedimentation rate and equilibrium in an ultracentrifuge.

In addition, some of these properties, especially viscosity and light scattering, provide valuable information on molecular shape. The viscosity and light scattering behavior of long rod-like molecules is quite different from that of compact sphere-like molecules of the same molecular weight. Further information on structure is, of course, obtainable from X-ray and neutron diffraction studies.

Carbohydrates

The carbohydrate polymers, or polysaccarides, of particular interest include cellulose, starch and pectin.

Cellulose is the principal structural constituent of plant life and is one of the most abundant of naturally occurring organic substances. It consists of a long linear polymer of a single six carbon sugar, d-glucose. The structure is shown below:



Starch

Molecular weight determinations by viscosity and osmotic pressure methods indicate that as many as one thousand glucose units may be linked in a single molecule. This linear polymer configuration is responsible for the familiar fibrous structures of naturally occurring cellulose materials. A bundle of long polymer molecules are held together in roughly parallel fashion by hydrogen bonds of the hydroxyl groups to form a fiber. The nature of hydrogen bonds will be discussed later in this chapter in connection with protein structure.

The principal effect of radiation on cellulose is a degradation produced by random bond rupture along the chain (Long and Lirot 1957). This has been determined by the decrease in the viscosity of solutions of irradiated cellulose. In addition to this chain scission, some of the glucose units are themselves destroyed, others are freed. The principal consequences of the chain scission are a loss in mechanical strength, an increase in the ease of hydrolysis and lowered resistance to bacterial attack.

Starch is the food reserve carbohydrate of all plants. It is, loosely speaking, also a polymer of d-glucose units, but consists of two distinct types of molecules. About 25 per cent consists of a linear glucose polymer called amylose and the remainder of a glucose polymer with side chains, called amylopectin. The amylose structure is very similar to the cellulose structure. The side chains of the amylopectin are substitutions of the hydroxyl hydrogen of the CH_2OH group. Amylose is a polymer of about 10,000 glucose molecules while amylopectin contains about 60,000 glucose molecules.

One of the well known color tests for starch is the iodine test. Addition of iodine to starch solutions produces a blue color. It has been found that this conspicuous blue color is due to amylose-iodine complexes, the amylopectin producing a violet or purple color. Spectrophotometric studies have revealed that the blue amylose color shifts towards the red as the molecular weight of this linear polymer decreases. When less than about 35 glucose units are combined the color departs from blue, at 8–12 units it is red and below this, the color disappears completely.

Irradiation of starch solutions produces depolymerization, as evidenced by reduced viscosity, color changes in the iodine complex, formation of reducing sugars and increased susceptibility to enzymatic hydrolysis. There is also evidence of some oxidation of the reducing groups to carboxyl groups. In Fig. 39A is shown the effect of radiation on the presence of high molecular weight amylose molecules, characterized by a light absorption peak at a wavelength of 660 mμ. It is seen that 1.5 megarads produce a substantial depolymerization of about half of the starch in a 2 per cent starch solution (Burns and Desrosier 1956). In Fig. 39B the rates of enzymatic hydrolysis of unirradiated and irradiated starch are shown. As the starch is hydrolyzed, there are fewer molecules available to form an iodine complex which absorbs at 660 mμ. It is seen that the rates of hydrolysis are parallel but displaced from one another due to the radiation-induced hydrolysis.

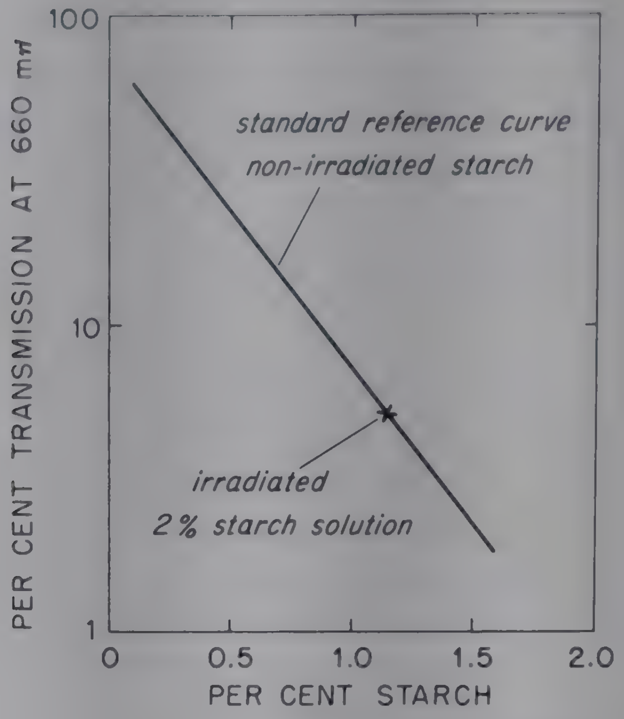
Table 22

RADIATION EFFECTS ON GLYCOGEN CONTENT OF GROUND BEEF[1]

Dose (megarads)	Glycogen (mg./g. meat)
0	0.44
2	0.26
4	0.17
8	0.13

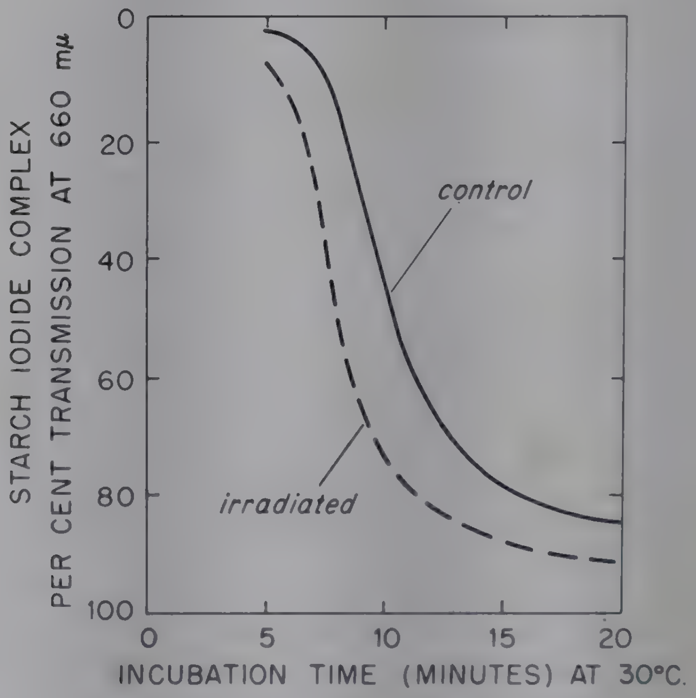
[1] From Schweigert and Doty (1958).

Pectin is a mixture of several carbohydrates. The principal one is galacturonan, a linear polymer which is similar to the cellulose structure. In place of the CH_2OH group branching from each ring, as in cellulose, there is a carboxylic acid grouping, COOH, or a methyl ester group, $COOCH_3$. Pectin is found in plant cell membranes and between cells, forming an adhesive which holds the cells together. It has an exceedingly high gelling power, being able to form firm jellies from fruit juices when present to the extent of only one per cent. When fruits and vegetables overripen the pectin gradually disappears and the decreased adhesion between cells in the plant structure produces a lessened firmness in the fruit or vegetable. Pectin is also depolymerized by



Courtesy of Desrosier and McArdle

FIG. 39A. EFFECT OF 1.5 MEGARADS ON IODOMETRIC PROPERTIES OF TWO PER CENT AQUEOUS STARCH SOLUTION BUFFERED AT pH 4.7 WITH SODIUM ACETATE



Courtesy of Burns and Desrosier

FIG. 39B. RATE OF ALPHA AMYLASE ACTIVITY UPON IRRADIATED AND CONTROL STARCH SOLUTIONS

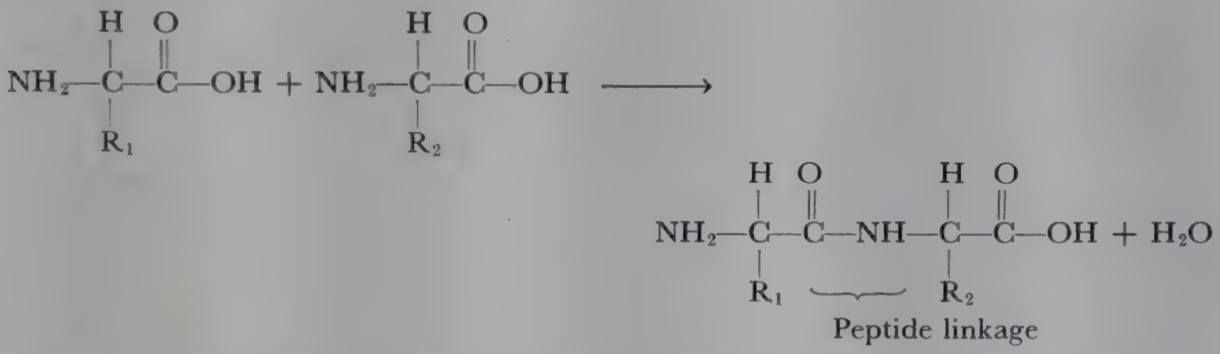
Rate of alpha amylase activity in 2.0 per cent starch solutions (*a*) irradiated with 1.2 megarads and (*b*) unirradiated control.

radiation, and is stated to be more radiation-sensitive than either cellulose or starch. However, it has been found that in the presence of sucrose, fructose or glucose sugars, the sensitivity towards radiation is much decreased. This is the case in jellies. In irradiated potatoes some weakening of the cellular structure has been observed which may be attributed to pectin destruction.

Glycogen is the reserve carbohydrate in animals. Studies on this substance in irradiated beef muscle indicate that some reduction in glycogen content results from irradiation as well. The glycogen content of beef flesh decreases with increasing radiation dose absorbed (Schweigert and Doty 1956). This is shown in Table 22. It is a common consequence of irradiation of animal flesh that it tastes sweeter than non-irradiated controls.

Proteins

Proteins are condensation polymers of the α-amino acids. The polymer units are held together by the so-called peptide linkage which is formed by condensation of the α-amino group with a carboxylic acid group of another amino acid (monomer) unit, splitting out a molecule of water.

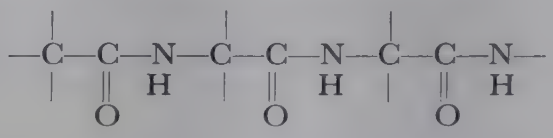


$$\mathrm{NH_2{-}\underset{R_1}{\overset{H}{C}}{-}\overset{O}{\overset{\|}{C}}{-}OH + NH_2{-}\underset{R_2}{\overset{H}{C}}{-}\overset{O}{\overset{\|}{C}}{-}OH} \longrightarrow \mathrm{NH_2{-}\underset{R_1}{\overset{H}{C}}{-}\underbrace{\overset{O}{\overset{\|}{C}}{-}NH}_{\text{Peptide linkage}}{-}\underset{R_2}{\overset{H}{C}}{-}\overset{O}{\overset{\|}{C}}{-}OH + H_2O}$$

In contrast to the carbohydrates discussed above, there is no simple repeating unit such as a glucose molecule. In naturally occurring proteins, the amino acids joined by peptide linkage consist of a varying number of some 21 amino acids. There are, then, to start with, three possible distinctions among various proteins—the amino acid composition, the sequential arrangement of the amino acids and the length of the polymer. This yields a spectrum of possibilities and, correspondingly, a great variety of proteins are known. Other substances such as hormones, enzymes, viruses and nucleoproteins are either proteins, strictly speaking, or are very closely related to them, differing mainly in the presence of characteristic side chain substitution groups (prosthetic groups).

While the amino acid composition is known for a number of proteins, the determination of the sequential arrangement of these building blocks is a much more difficult problem. In recent years chemical methods have been developed by Sanger (1956) and others which are beginning to supply answers to this question. To date, the most complex amino acid sequence which has been completely determined is that of the hormone insulin, a polymer of molecular weight 5733 containing 51 amino acid units of 16 different types. The magnitude of the problem is indicated by the fact that this molecular weight is only at the lowest end of the scale for proteins. Some proteins have molecular weights as high as eight million (crystallizable viruses).

The most common configuration of protein molecules known to date is the α-helix configuration (Newberger 1958). In this configuration the successive peptide linkages which form the backbone of the structure

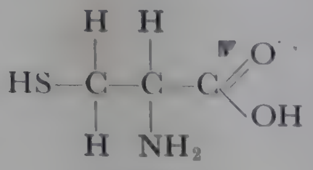


$$-\overset{|}{\underset{|}{C}}-\underset{\underset{\textstyle O}{\|}}{C}-\underset{H}{N}-\overset{|}{\underset{|}{C}}-\underset{\underset{\textstyle O}{\|}}{C}-\underset{H}{N}-\overset{|}{\underset{|}{C}}-\underset{\underset{\textstyle O}{\|}}{C}-\underset{H}{N}-$$

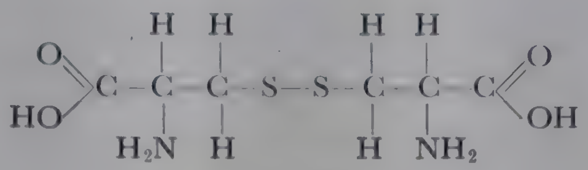
are arranged in a helical configuration in which about 3.5 linkages complete a turn in the helix. The substituents, R, which characterize the amino acids, are free in this structure to extend beyond the sides of the helix (see Fig. 8, p. 31). This helical configuration is consistent with the bond angles required of the peptide linkage itself.

The helical structure is also stabilized by a variety of secondary chemical bonds, of which an important one is the hydrogen bond. It is found that a hydrogen atom attached to an electronegative atom such as fluorine, oxygen, or nitrogen can form a weak bond with another such atom as the latter, two to three Angstrom units away. This bond is primarily electrostatic in nature and results from the circumstance that an electronegative atom will attract electrons to itself in a chemical bond. As a result of this, the electronegative atom carries a net negative charge equal to some fraction of a single electronic charge. The hydrogen, covalently bonded to such an atom, carries a net fractional positive charge. Thus, an electrostatic attraction can occur between the hydrogen and another electronegative atom. The strength of these hydrogen bonds is of the order of five kilocalories per mole, which is about 5 to 10 per cent of the strength of ordinary chemical bonds. In the helical structure it is seen that such a hydrogen bond is established between every NH hydrogen and every C=O oxygen. If a single protein molecule were to be uncoiled, the rupture of a large number of these comparatively weak bonds would require a considerable energy input.

Another important secondary linkage in almost all proteins is the disulfide linkage. The amino acid cysteine has a sulfhydryl group, SH, on the β carbon. It can combine in an oxidation reaction with another sulfhydryl group to form a disulfide linkage. This "dimerized" acid is called cystine. The hydrogen of the sulfhydryl group is oxidized to water.



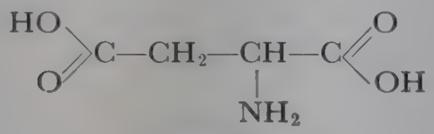
Cysteine



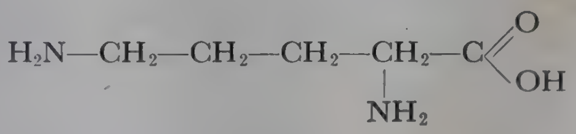
Cystine

In the protein insulin such disulfide linkages have been established between two cysteine units separated by a number of intervening amino acids. This would require a folded helical configuration in order to bring the two sulfur atoms close enough together to form a disulfide bond. Further, it has been determined that insulin consists actually of two adjacent helices containing 21 and 30 amino acids, respectively. The helices are held together to form a single insulin molecule by two disulfide bonds and probably some hydrogen bonds. It is to be noted that, except for hydrogen bonds, the disulfide bond is one of the weakest bonds found in proteins (~ 20 kcal).

Another feature of proteins is their acid-base characteristics. A number of the 21 α-amino acids of which most proteins are built consist of α-amino acids which have a substituent group that is itself acidic or basic. Examples are shown below.



Aspartic Acid
(acidic)



Lysine
(basic)

Depending on the amino acid composition, the protein can act as an acid and/or base with a whole sequence of dissociation constants, much as phosphoric acid H_3PO_4 can dissociate to form, in turn $H_2PO_4^-$, $HPO_4^=$, and $PO_4^{\equiv}$ ions. An important consequence of this is that a protein molecule in solution can be a multiply charged cation or anion. And the amount of charge can be made to vary by varying the pH of the solution. There is for every water-soluble protein a unique pH value at which the molecule has no net charge, the acidic and basic dissociation being just equal. This is called the isoelectric point. At a pH away from the isoelectric point the protein is a large ion with a pH dependent charge, and some properties of this ion can be determined by causing it to move in an electric field. This experimental technique is called electrophoresis, and is in many ways analogous to electrolytic conductance experiments. The technique is a powerful tool for studying the purity of protein preparations, which frequently are prepared from material containing not only other substances but frequently a number of different proteins. The electrophoretic behavior is generally characteristic for each protein at any given pH.

An important characteristic reaction of proteins (and protein derivatives) is protein denaturation (Kauzmann 1954). Perhaps the most familiar example of protein denaturation is the conversion of raw egg white into rigid white egg white by heat. There is unfortunately no simple clearcut definition of denaturation. We adopt the definition that denaturation is any change in a physical or chemical property of the protein that can be brought about without a change in the molecular weight of the protein. Historically, the first criterion for denaturation was the conversion of a soluble into an insoluble protein in a given solvent, for example the egg white transformation. This effect involves coagulation of protein molecules into larger, insoluble aggregates, a step which appears directly to violate the above definition. There is good evidence, however, that the coagulation step is a secondary step incidental to a prior profound change in the protein molecule.

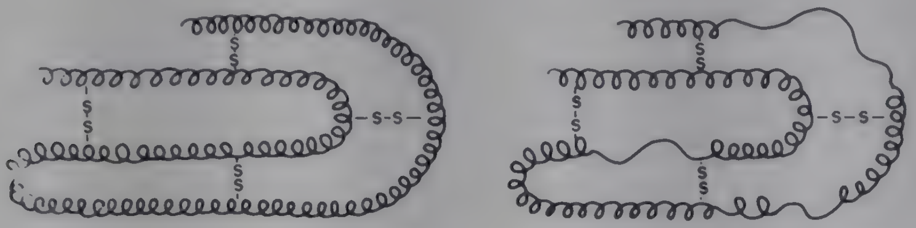
Protein properties which may change in the denaturation process include:

1. Solubility
2. Optical rotation
3. Ultra-violet absorption spectra
4. Light scattering
5. Viscosity
6. Acid-base characteristics
7. Change in number of characteristic substituent groups accessible to chemical reaction, particularly S-H groups
8. Enzymatic activity (in enzymes)
9. Susceptibility to enzymatic attack

The agents which can bring about denaturation include:

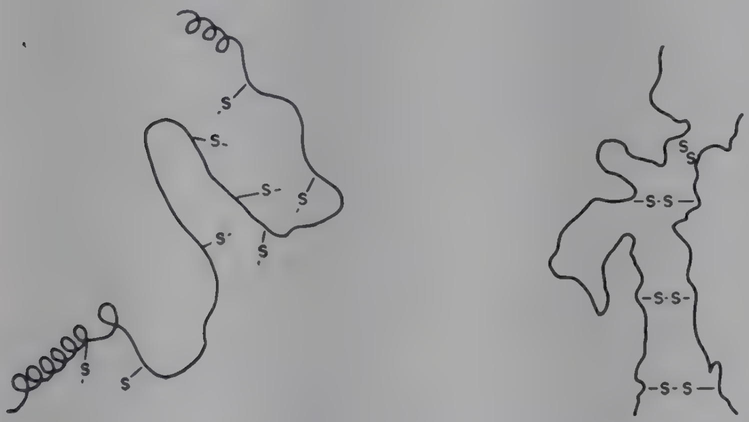
1. Radiation
2. Heat, pressure, freezing, ultrasonic waves
3. Chemical agents such as acids and bases, organic solvents such as alcohol, urea and other amides and detergents
4. Enzymes

The one hypothesis which correlates a large part of the information on denaturation is that this process involves a change in the structure of the protein molecule. A typical molecule in the native (undenatured) state is, as we have seen, essentially a helical structure which is sometimes folded on itself. A denatured protein may be visualized as the same molecule in a more or less unfolded configuration. The limiting case would be a random coil, Fig. 40. It has been found that the viscosity of denatured ovalbumin and serum albumin is comparable to that of random coil synthetic polymer of the same chain length. The changes in optical rotatory power on denaturation are also in accord with this picture. It is evident that in a tightly folded, coiled protein molecule some chemical groups may not be accessible to reaction. Further, even small structural changes such as partial uncoiling can have drastic effects if the substance is an enzyme. Not much change is needed to destroy enzyme activity (see below).



A. Native
(coiled, folded)

B. Reversibly denatured
(partially uncoiled, folded)



C. Irreversibly denatured
(uncoiled, unfolded)

D. Coagulated protein

Fig. 40. Schematic Diagram of a Protein Molecule in Various Stages of Denaturation

With the above model (very much simplified) of a protein and the denaturation criterion, the effect of some of the denaturing agents can be understood. When an acid or base is added to an aqueous protein solution, a shift in the ionization of the basic or acid groups occurs. In effect, one has done a titration. The protein molecule now bears an excessive number of negative or positive charges. The electrostatic repulsion between these charges disrupts the structure. The organic solvents have one common characteristic, they are hydrogen bond donors (ROH,

RNH_2) or acceptors ($R_2C{=}O$). These substances compete with the internal hydrogen bonding in the protein helix and, by offering alternate bonding partners, allow the helix to uncoil. The detergents likewise compete similarly for secondary bonding in the protein. The effect of heat is probably connected with the temperature dependence of the effectiveness of water as a hydrogen bond competitor in the helix. A similar explanation probably holds, in part, for pressure induced denaturation. In this case, however, another factor is that denaturation involves a volume change in the protein molecule, sometimes positive, sometimes negative. The pressure effect is no doubt partly due to this factor. It is found that sometimes a high pressure makes denaturation more difficult.

An important question is whether denaturation is a reversible process. The case of egg white would patently say no. On the other hand, an enzyme, chymotrypsinogen, can be reversibly denatured (inactivated) by heat. Present evidence tends to favor the statement that every denaturation is reversible up to a point, after which an irreversible process such as coagulation follows. Further evidence in favor of this view is that recent studies on optical rotation of aqueous protein solutions are consistent with the picture that in a protein solution there are found all sorts of species from completely ordered (native) protein through more or less uncoiled to completely uncoiled unfolded species (Doty 1959). This is in accord with an equilibrium and a reversibility of the process.

Effects of Radiation

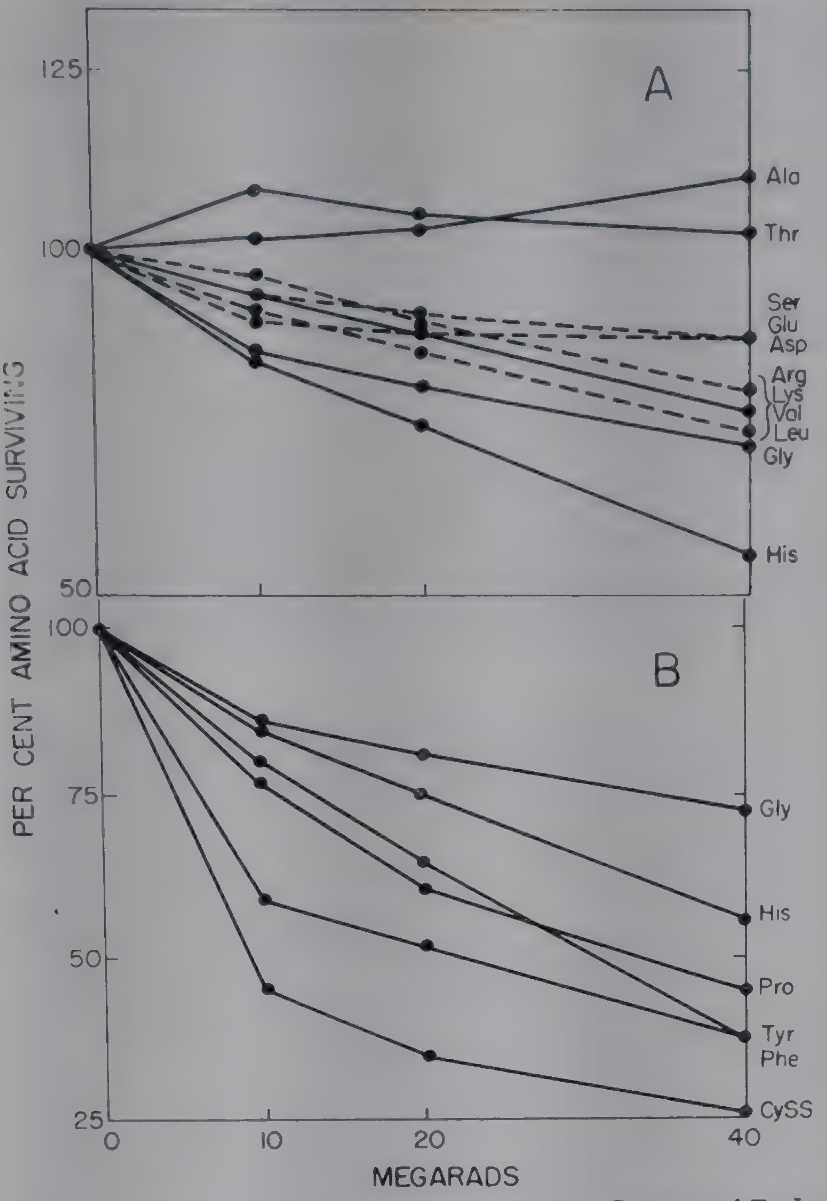
The effects of radiation on proteins can be divided into three classes:

a. Specific chemical changes

b. Changes in physicochemical properties, including denaturation

c. Less clearly defined changes, particularly odor formation, which are of technological interest.

The demonstrable chemical changes in irradiated proteins include liberation of ammonia, oxidation of S–H groups and formation of peroxides (Drake and Giffee 1957). The ammonia may be liberated from free NH_2 groups which occur at one end of the molecule, from NH_2 groups which are not an integral part of the protein backbone (as in a dibasic amino acid), and from ruptured peptide linkages. This behavior parallels that found in aqueous amino acid solutions see Table 17, p. 129). The oxidation of S–H groups is inferred from changes in enzyme activity. This oxidation probably occurs by attack of an OH radical formed in water. Another effect on sulfur in proteins is the liberation of H_2S. This, again, parallels the behavior of single amino acids. The nature of the peroxides formed on irradiation is not known, but the presence of this



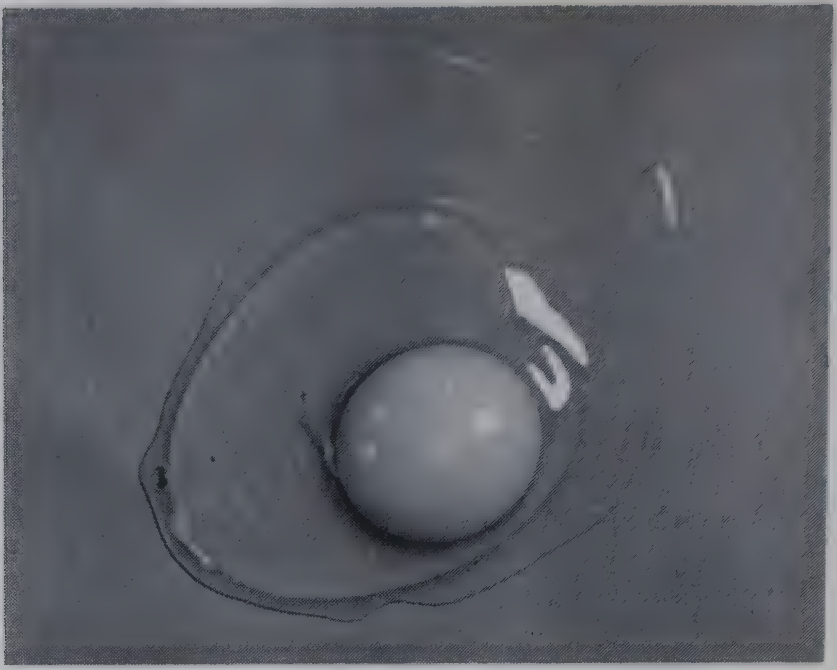
Courtesy of Drake

FIG. 41. PER CENT OF AMINO ACID SURVIVAL ON ONE PER CENT INSULIN AS A FUNCTION OF IRRADIATION DOSE

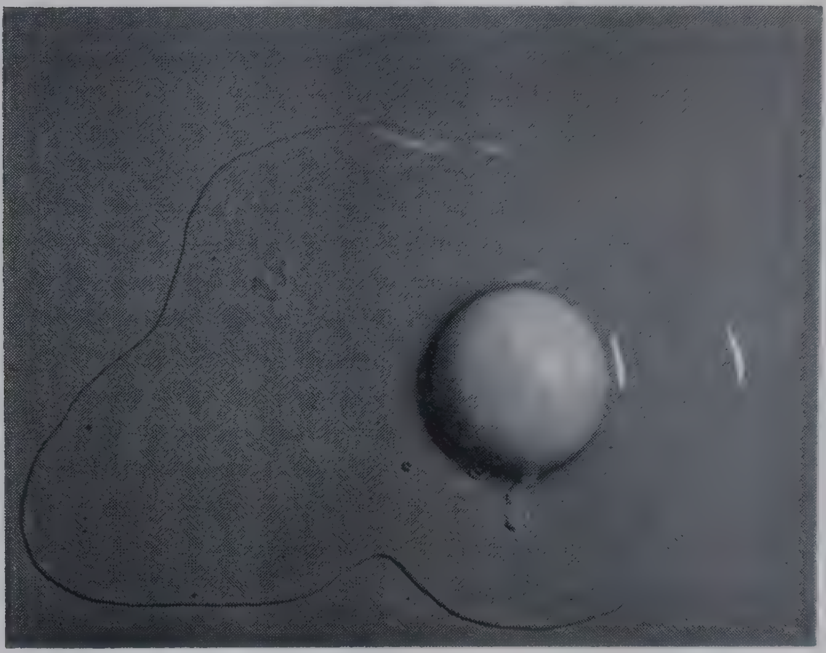
Amino acid control used as 100 per cent

effect should be expected from considerations given in the preceding chapter.

A protein molecule behaves in the presence of radiation both as a single molecular entity when it is denatured, and also as an assembly of various amino acids which each display a characteristic radiation sensitivity. In the case of insulin, whose composition is known, the relative degree of destruction of the individual amino acid components has been determined by Drake *et al.* (1957), see Fig. 41. It is seen that various components have different sensitivities, with the sulfur containing cysteine, CSS, the most sensitive. Further, there is an initial increase in the amount of threonine, CH_3 CHOH $CHNH_2$ COOH, which may result from radiation-



Control



600,000 rads

Courtesy of McArdle and Desrosier

FIG. 42. RADIATION EFFECTS ON FRESH EGG PROTEIN

With 600,000 rads, there is disappearance of thick albumin

induced chemical transformation of one of the other components, possibly valine, $(CH_3)_2CHCH_2NH_2COOH$.

The most characteristic reaction which an irradiated native protein undergoes is denaturation. This can be clearly seen in a very subtle demonstration in Fig. 42. An egg consists of a yolk suspended and held in position by chalaza in a water suspension of albumin. The albumin in freshly laid eggs contains a thick and a thin component. The thickness of the albumin shown in the figure is due to the presence of fibers of the protein ovomucin. The structure of this thick albumin, held by ovomucin fibers, is most fragile, being altered even by agitation or stirring. Its destruction by irradiation is clearly seen. Ordinary chemical analyses at best yield minute differences between the irradiated and control samples (McArdle and Desrosier 1955).

The following general observation may be made about radiation induced protein denaturation. Radiation produces changes in viscosity, ultra-violet absorption spectra, electrophoretic behavior, altered sensitivity towards enzymes, increase in exposed SH groups and coagulation or precipitation. These changes are characteristic of denaturation. Further, an irradiated protein is more sensitive towards subsequent heat denaturation, although prior heat treatment does not increase sensitivity towards radiation denaturation. This effect is useful in sensitizing harmful bacteria for heat destruction, as will be described in Chapter 12. The above types of changes have been demonstrated with doses as small as 33,000 rads and, as expected, differ from protein to protein.

Table 23

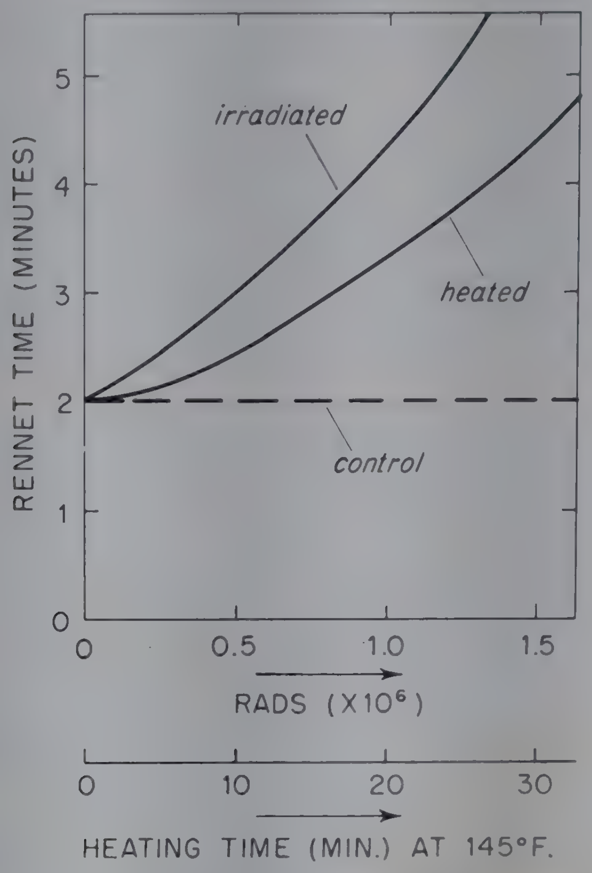
THE LIBERATION OF SULFHYDRYL GROUPS IN IRRADIATED AQUEOUS PROTEIN SOLUTIONS[1]

	—SH as micrograms cysteine·HCl per ml.	
Protein	Control	1.5×10^6 rads
2 per cent casein	0.22	3.77
Ascorbic acid (0.5 mg./ml.)	0.25	1.82
2 per cent albumin	0.44	9.21
Ascorbic acid (0.5 mg./ml.)	0.44	3.17

[1] From McArdle and Desrosier (1955).

Several mechanisms have been proposed to account for radiation-induced denaturation. The first, by Augenstine (1958), focuses attention on the rupture of S-S bonds in the protein as a result of transfer of excitation energy into these bonds after the passage of radiation through the molecule. A second mechanism, due to Platzman and Franck (1958), attributes the denaturation to rupture of hydrogen bonds as a consequence of the distortion of the molecular configuration by the electric charge distribution created in the molecule by the passage of ionizing radiation. The latter mechanism appears preferable. However, it is not sufficient to account for denaturation of protein monolayers by electrons of very low energy, less than the ionization potential of a molecule. There are also examples of protective action of various substances in retarding protein denaturation by radiation (Dale *et al.* 1949). This is particularly true of substances containing SH groups such as thiourea and B.A.L. (British Anti-Lewisite) which contains many SH groups. Ascorbic acid, which contains no SH, also exhibits some protective effect on casein in the presence of irradiation. See Table 23.

Two proteins of particular interest in food irradiation are casein, a constituent of milk, and egg albumin.



Courtesy of McArdle and Desrosier

FIG. 43. COMPARISON OF RENNET COAGULATION TIMES FOR IRRADIATED AND HEATED MILK

The reduction in thickness of natural egg white by ionizing radiations initially is opposite to the effect on the relative viscosity of purified albumin in aqueous solution, irradiation resulting in an increased relative viscosity of the latter.

The results of experiments on irradiation of systems of casein and egg albumin indicate that in both proteins there is an unfolding of the molecules. The pattern followed differs in the two proteins, but the increase in free sulfhydryl groups demonstrates conclusively that molecular rearrangement has occurred. Since this molecular change does not result in any marked increase in amino nitrogen it is apparent that the peptide linkages are not the major site of attack. The sulfur linkages are very definitely the site of a large share of the radiation effect, hydrogen bond linkages also being vulnerable.

Ionizing radiations produce changes in native milk casein which result in an increase in its coagulation time by the rennet enzyme and a reduction of its stability to heat.

Similarities between the effects of radiation and of heat on milk are apparent in the influence of each on the rennet coagulation time. In both cases there is a decrease in rate of enzyme activity (Fig. 43).

The influence of the addition of calcium on the rennet coagulation of irradiated or heated milk is significant. The effect indicates that in both treatments calcium is precipitated as tricalcium phosphate, a form in which it cannot function in the action of rennet. This precipitation of calcium is also of importance in the stability of casein to heat, acid, or other denaturing substances. Irradiation produces an effect on the calcium balance which is very similar to that which results from the heat treatment. The available calcium is reduced. It combines with activated protein molecules in such a way as to be inactive in the rennet reaction or as a stabilizing element. Considering the excitation and ionization reactions which occur in irradiated substances it is not difficult to visualize the calcium becoming combined into a complex of protein molecules. This seems more probable in the light of the results obtained in studying the effects of cathode rays on purified casein in aqueous solution. Here there is evidence based upon changes in relative viscosity of the solution, of association of casein molecules or fractions of molecules. The calcium is easily combined in such reactions. If the element is added before treatment (irradiation or heat) it does not affect the rennet time after treatment. The surplus calcium is also made unavailable.

The changes in flavor resemble those which occur when milk is heated. Taste panel results indicate some similarity between the off flavor of irradiated milk and the burnt flavor of over-heated milk.

Radiation Effects on Enzymes

Enzymes are biological catalysts, in their presence complex biochemical materials can undergo rapid, specific transformations which still defy conventional chemical techniques. They are an essential part of the chemical machinery of living material, although they can carry out their function in isolated form. The digestion of carbohydrates and proteins—and their synthesis—as well as respiratory processes are examples of chemical transformations in living materials which are controlled by enzymes. Each of these transformations is a complex sequence of individual chemical steps and, correspondingly, with each of these transformations there is associated an entire coordinated set of enzymes each of which exerts its function in one of the chemical steps. The number of enzymes which have been isolated runs well into the hundreds. Of these, over one hundred have been isolated in crystalline chemical form. Their molecular weights, insofar as they have been determined, run upwards of about twenty thousand.

In view of the complexity of the problem—high molecular weight materials catalyzing specific changes in complex biochemical substances—it is not surprising that the details of the structure and function of these substances are only beginning to be understood, after many years of thorough and inspired research. And one must approach generalizations with due caution. However, two generalizations appear valid, one on structure and one on functionality, which provide a basis for a qualitative understanding of radiation effects on these materials (Neurath 1959).

So far as is known, all enzymes consist basically of proteinaceous material. In some cases, chemical analysis indicates that the functional enzyme consists exclusively of amino acids; in other cases there is conclusive evidence that with this protein matrix there are associated one or more substituent groupings (prosthetic groups), consisting of complex organic substituents, metallic ions, or both which are essential to the catalytic role of these materials.

The specificity of the enzyme is accounted for by the postulate that a given enzyme bears a particular number of active sites, organized in a definite spatial relationship, to which the substrate molecule must be held in order to undergo a chemical transformation. The forces holding the molecule to the active sites are conventional chemical forces, such as hydrogen bonds or other electrostatic forces. As was seen earlier, individual chemical groupings have different bonding power to a particular grouping, or site. Thus it appears reasonable to suppose that between the active groupings of the enzyme and the material acted upon—the substrate—there must exist a degree of compatibility of chemical groupings and their spatial arrangement, in order that the substrate molecule can be physically bound to the enzyme for subsequent chemical transformation. This relation has been aptly described as a "lock and key" relation. The details of the molecular events following this attachment, which produce the chemical change and subsequent detachment, are still obscure.

The lock-and-key metaphor may profitably be carried a little further. Within limits, there is some freedom concerning both the shape of the lock housing and the shape of the handle of the key. It is found, on the one hand, that a particular enzyme can still carry on its function, in vitro, even if one end of the enzyme is removed—up to a point. For example, three amino acids can be removed from one end of the enzyme chymotrypsin but not four, without destroying enzymatic activity. Further, enzymes are specific but only up to a point. Proteolytic enzymes each can cleave peptide bonds between a rather restricted set of amino acid pairs, and can do so with partial disregard of the molecular structures depending from the other ends of the amino acid pairs. However, a particular amino acid pair in a protein may be far more susceptible to lysis by a given enzyme if the protein molecule is denatured than it is if the protein is in the native (or undenatured) state. There is little doubt that the complete elucidation of enzyme action will reveal a wide spectrum of bonding interaction, spatial relationships and molecular mechanisms of chemical change.

The importance of the spatial relationships of the enzyme and the substrate, together with the basic protein structure of enzymes suggests a close parallel between enzyme inactivation and protein denaturation. And this is indeed found. It was mentioned earlier that protein denaturation consisted mainly of alterations in the configurations of protein molecules. It is clear that a sufficiently profound alteration in the configuration of an enzyme molecule would destroy the conformation between the arrangement of active sites and substrate. It is, in fact, found that the chemical and physical methods of protein denaturation will very frequently inactivate an enzyme. And further, in some cases the inactivation is reversible, as is the denaturation. This suggests that the mechanism of radiation denaturation and radiation inactivation are very closely related, at least in the case of enzymes composed exclusively of amino acids.

In contrast to studies on proteins, the effects of radiation on enzymes are generally measured in terms of the loss in enzymatic activity. In view of this, no particular significance can be attached to straight comparisons between protein and enzyme radiation sensitivity. For example, it was seen that the removal of a small number of amino acids from one end of an enzyme molecule did not affect its activity, whereas the removal of one more acid destroyed it. If this were the result of an irradiation, this effect could be readily detected with the enzyme but might escape notice in a protein molecule for which the measure of damage was arbitrarily chosen to be denaturation.

Many studies on enzyme irradiation have been carried out on different systems under a variety of conditions. These can be grouped into three general categories:

a. Irradiation of pure, dry, crystalline enzymes
b. Irradiation of aqueous solutions of pure enzymes
c. Irradiation of enzymes in real environments such as cells, cell homogenates, food components and equeous solutions containing a small number of specific additives.

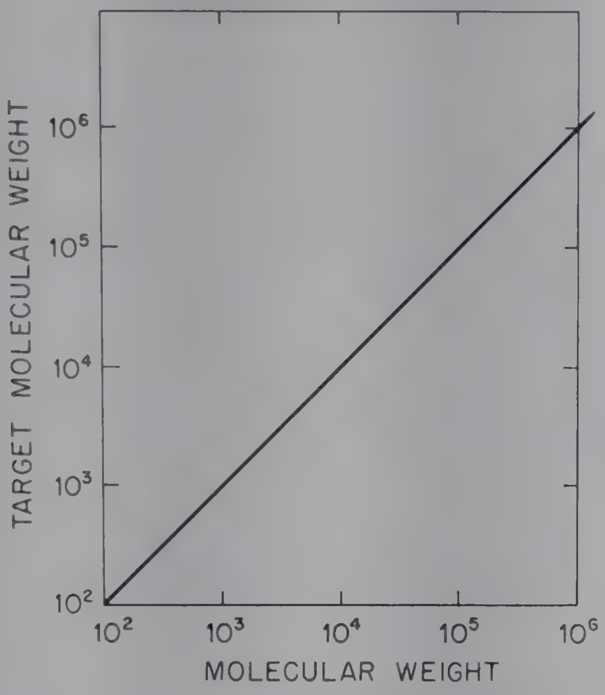
This variety of conditions and the great variety of enzymes studied obviously represent a large number of possibilities for all varieties and gradations of radiation effects. And these are, in fact, found. However, the following may be offered as provisional generalizations in this area.

Among the three sets of conditions, the pure aqueous solutions of enzymes are by far most radiation sensitive. Doses of as little as several hundred rads can produce pronounced destruction of enzymatic activity (several per cent or more). In contrast, a corresponding effect in crystalline enzymes, or in real environments—excepting aqueous solutions with additives—requires doses of the order of a megarad. Aqueous enzyme solutions containing one or more specific additives are frequently as radiation resistant as enzymes in cells or foodstuffs, depending on the additive. This indicates that a number of additives can function as very effective protective agents. In the case of crystalline enzyme preparations, it is found that enzymes become more radiation sensitive as their molecular weight or size increases (Pollard 1959). In this respect, as in others to be mentioned, enzymes appear to represent to radiation a simple geometric target which can be destroyed by one or a small number of direct hits. This description of radiation effects is the famous target theory, developed by Lea (1956) and others. It has been applied both to complex biochemical molecules such as enzymes and to living cells.

The assumptions of the target theory may be stated in simplified form as follows. In order for a chemical or functional transformation to occur in a complex biochemical molecule, it is necessary and sufficient that ionization and/or excitation be directly produced in the molecule. Further, in most cases the occurrence of a single such event in the molecule is sufficient to bring about this transformation. In the case of cells, in regard both to gene mutations and to mortality, it is assumed that the ionization and/or excitation of a critical component molecule, such as a gene, is necessary and sufficient to bring this about. In the case of some radiation induced effects it is necessary that a small number of such events occur at a particular molecular site. The assumptions of this theory specifically exclude the possibility that active agents produced outside the cell or molecule can diffuse to the target and bring about the observed transformation.

This theory has a number of verifiable consequences. If a single ionizing or exciting event suffices to bring about this change, the number of survivors decreases exponentially with dose and if more than one event is required the survivor curve assumes one or another sigmoidal shape. Further, the amount of transformation should be essentially independent of dose rate in the single event case. If the quality of the radiation changes from sparsely ionizing (gamma) radiation to densely ionizing (alpha) radiation, the dose required to bring about a given amount of change increases because of unnecessary multiple ionization of a single target. Finally, the spatial distribution of ionization is sufficiently well known for high energy radiation to permit calculation of the target size which is required to account for the observed transformation, and this target size must be in accord with reality.

The above consequences of the target theory have been verified in many cases, including cell mortality, genetic mutation, and destruction of viruses, enzymes and proteins in the pure crystalline state. In the case of enzymes, exponential survival curves are found. From the target size a molecular weight may be calculated by assuming reasonable values for the density of enzyme molecules, and the results are in impressive accord with other molecular weight determinations. A particularly striking relation is shown in Fig. 44 between the target molecular weight and the molecular weight determined by other means for a number of hormones, enzymes, proteins and viruses (Pollard 1959).

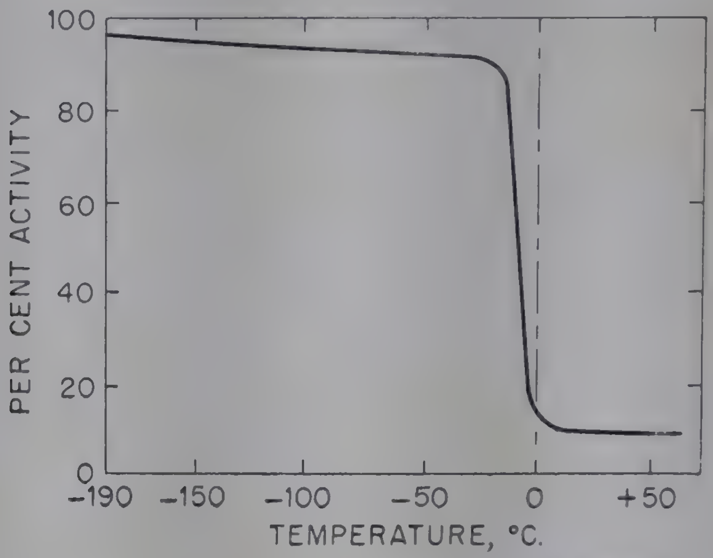


Courtesy of Pollard

FIG. 44. A PLOT COMPARING THE OBSERVED MOLECULAR WEIGHT FOR A WIDE VARIETY OF BOMBARDMENT SUBSTANCES

As indicated above, the inactivation doses for enzymes in aqueous solutions are very low, doses of several hundred rads leading to substantial inactivation in some cases, for example, for plant growth hormone synthesis. It is of interest to note that irradiation of the proteolytic enzyme papain produces an *increase* in enzymatic activity at doses of 100 rads and less, followed by a decrease at higher doses.

In a number of enzyme solutions it is found that the specific radiation sensitivity decreases as the (small) concentration of enzyme increases. In the case of pepsin, the dose required for 63 per cent inactivation of a solution of 10 mg/ml concentration is about two and one half million rads, compared to only about 40 thousand rads for a solution of one hundredth the above concentration (Lea 1956). This concentration effect indicates that in aqueous solution, at least, the simple target concept is inadequate. Further evidence is found in the fact that a number of substances added to aqueous enzyme solutions exert a more or less pronounced protective effect on radiation inactivation. Such protective substances include, in the order of decreasing protectiveness, thiourea, sodium formate, dimethylthiourea, glucose and egg albumin. In contrast to thiourea, the most effective protective agent, ordinary urea is less effective by a factor of about 5,000. On the basis of the not too plentiful data extant it appears that some protective agents appear to function in the presence of a wide variety of enzymes; other agents may be quite specific to a particular enzyme. The general features of protective action of such additives clearly indicate that in irradiated aqueous enzyme solutions one or more active species are produced in the water which are capable of inactivating enzyme molecules. In other words, inactivation can occur by mechanisms other than a "direct hit" as assumed by target theory. Further, in these aqueous systems exponential survival curves are frequently found. This indicates that the exponential survival curve by itself is not itself diagnostic of a target effect. Finally, the protective action of the protein egg albumin suggests that the decreased radiation sensitivity of enzyme solutions at higher concentrations is a manifestation of protection on the part



Courtesy of Bellamy

FIG. 45. EFFECT OF TEMPERATURE ON THE INACTIVATION OF 10 MG. PER ML. SOLUTION OF PEPSIN BY 2,500,000 RADS

of inactivated enzyme molecules which, it will be recalled, are themselves largely protein structures.

The radiation sensitivity of enzymes increases as the temperature is raised. This has been found both for enzyme solutions and crystalline preparations (Siu 1957). This behavior parallels that found in proteins undergoing radiation denaturation. When enzyme solutions are frozen, the radiation sensitivity is decreased considerably, probably because of the immobilization of active species produced in the surroundings (see Fig. 45). The greatly decreased sensitivity, a factor of ten in an aqueous pepsin solution, gives some indication of the comparative importance of direct action (target) and indirect action.

The radiation inactivation of enzymes in cell homogenates, tissues and foodstuffs is of considerable technical importance. It is evident that the end result of irradiation follows from a complex sequence of events including direct inactivation by direct hits, indirect inactivation by externally generated active species and the protective, and perhaps sensitizing, action of the enzyme surroundings. For a wide variety of enzymes, inactivation is not serious at doses up to one megarad and, as expected, varies from one enzyme to another, see Table 24. Further, it is found that the sensitivity of a particular enzyme depends on the nature of the substrate, as shown for the beef proteolytic enzyme in Table 24. This variation probably reflects the differences in protective action of the components of the different tissues, and the subtle differences in these proteolytic enzymes.

Table 24

INACTIVATION OF FOOD ENZYMES BY IONIZING RADIATIONS[1]

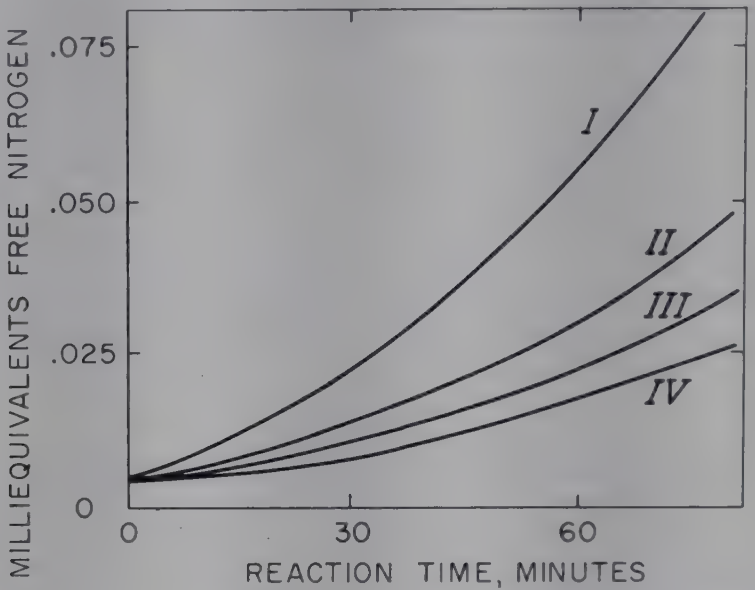
Enzymes	Dose in Megarep	Inactivation in Per cent
Alkaline phosphatase in milk	5	92.3
Succinoxidase in beef liver	1	64
Succinoxidase in beef liver with 60 gammas of cytochrome c added	1	10
Proteolytic activity of beef spleen	2	51
Proteolytic activity of beef kidney	2	43.5
Proteolytic activity of beef heart	2	29
Mushroom tyrosinase	2	37
Cucumber ascorbic acid oxidase	2	25

[1] From Siu (1957).

Enzyme reaction rates have been studied regarding the effect of irradiation upon the enzyme, the substrate, and their combination, on the resulting activity (Fig. 46). A radiation dose which does not destroy the enzyme in a suitable substrate finds the reaction proceeding. If an untreated enzyme is allowed to react on an irradiated substrate, the reaction rate is increased, when compared with a non-irradiated substrate. The increase generally occurs, and perhaps is due to an activation of the substrate. With irradiated protein, ionization apparently causes an unfolding of the protein molecule, rendering the points of attack more accessible to the enzyme (McArdle and Desrosier 1955).

The radiation resistance of enzymes varies, as demonstrated in Table 24. Resistance is dependent on the temperature of irradiation, concentration of enzyme, pH of substrate, presence of moisture, and free radical acceptors. The inactivation dose decreases with increasing temperatures.

Oxygen effects have not been shown. Radiation resistance of dry enzymes are in the realm of 100 times greater than in water solutions.



Courtesy of McArdle and Desrosier

FIG. 46. EFFECTS OF IRRADIATION ON ENZYME—SUBSTRATE REACTION RATES (ENZYME—TRYPSIN; SUBSTRATE—CASEIN; DOSE —1.5 MEGARADS)

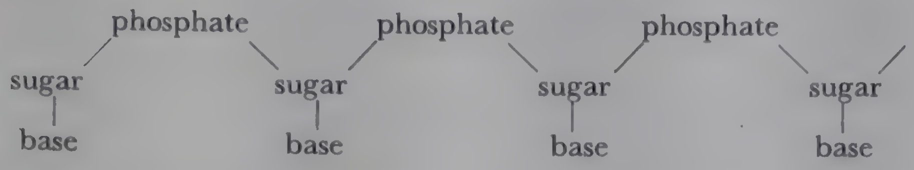
I—enzyme control, substrate irradiated; *II*—enzyme control, substrate control; *III*—enzyme irradiated, substrate irradiated; *IV*—enzyme irradiated, substrate control

The relatively low sensitivity of enzymes to radiation effects is of practical interest in foodstuffs because it is necessary to prevent enzymatic spoilage as well as microbial spoilage in preserving food. The inactivation of enzymes by moist heat usually coincides with the thermal destruction of vegetative forms of organisms. This is not the case with radiation preservation of foods. As a general rule, the complete inactivation of enzymes requires in the order of five times the dose required for the destruction of micro-organisms.

Because of the low sensitivity of enzymes to radiation, and because the substrates can be rendered more susceptible to attack by enzymes, while at the same time failing to inactivate them, radiation sterilization can bring about a rapid deterioration in killed plant and animal tissues unless enzymes are controlled.

Radiation Effects on Nucleoproteins

The nucleic acids are biological polymers which are important in the transmission of genetic information. Their molecular weight is of the order of several million. The polymers consist of a backbone of alternating five-carbon sugar and phosphate groups. A variety of organic bases is attached to this backbone, one at each sugar group.



There are two classes of nucleic acids. In one class, DNA, deoxyribose is the sugar part of the backbone, and in the other class, RNA, ribose is the sugar part of the backbone. The organic bases are four in number, adenine, quanine, and cytosine being common to both classes and the fourth member being uracil (thymine) in RNA and 5-methyl-uracil in DNA.

In recent years a considerable amount of information has been accumulated about the molecular configuration of DNA, whereas the information on RNA is still quite fragmentary. The present day picture of DNA is a helical structure in which two strands run antiparallel to one another; it is, thus, a double helix. The two strands are held together by hydrogen bonds between the bases. The helical structure of this polymer imparts a high viscosity to aqueous nucleic acid solutions.

Irradiation of pure DNA in the crystalline state destroys its biological effect on pneumococci. The results suggest that crystalline DNA presents two sets of targets to radiation, having a molecular weight of 5 to 15 million and 300,000 respectively (Pollard 1959). The reason for this difference is not understood, it may possibly represent two mechanisms of radiation deactivation.

Many studies have been performed on aqueous DNA solutions and in almost all of them the measure of effect is the change in viscosity. Irradiation produces a decrease in viscosity at doses of several million rads and higher, the reduction being greater at higher doses. In other studies it is found that doses as small as 5,000 rads produce a decrease in the viscosity in pure DNA solutions and still lower doses can affect the viscosity of the DNA associated with a protein. There is as yet no clear interpretation of the viscosity changes. Further observations are that a variety of substances such as glucose, methanol, serum albumin and thiourea exert a protective effect. Also, there is an irradiation after-effect, the viscosity continuing to decrease for several hours after irradiation. Chemical tests show that irradiation of DNA solutions produces some oxidation of the sugar groups, deamination, liberation of free bases and phosphate groupings, introduction of hydroperoxide groups and changes in the ultraviolet absorption spectra (Butler 1959). It is clear that these changes cannot be simply related to radiation-induced genetic change, the amount of change required for a genetic effect is probably undetectable by present-day chemical or physical methods.

BIBLIOGRAPHY

Augenstine, L. G. 1958. A proposed mechanism of protein inactivation. In Yockey, H. P., Platzman, R. L., and Quastler, H., Eds., Symposium on Information Theory in Biology, 287–292. Pergamon Press, New York, N. Y.

Barron, E. S. G. 1954. The role of free radicals and oxygen in reactions produced by ionizing radiations. Radiation Research *1*, 109–124.

Bacq, Z. M., and Alexander, P. 1955. Fundamentals of Radiobiology. Academic Press, Inc., New York, N. Y.

Bernstein, M. H. 1954. Deoxyribonucleoproteins of cell nuclei. Sensitivity to X-rays. Nature *174,* 463–464.

Bourne, E. J., Stacey, M., and Vaughan, G. 1956. Action of gamma radiation on dilute aqueous solutions of amylose. Chem. and Ind. June 30, *11*, 573.

Bier, M., and Nord, F. F. 1951. The effect of certain ions and of radiation on crystalline trypsin. Arch. Biochem. and Biophys. *31*, 335–337.

Bovey, F. A. 1958. The effects of Ionizing Radiation on Natural and Synthetic High Polymers. Interscience Publishers, New York, N. Y.

Burns, E. E., and Desrosier, N. W. 1956. Maturation changes in tomato fruits induced by ionizing radiations. Food Technol. *11*, 313–316.

Butler, J. A. C. 1959. Changes induced in nucleic acids by ionizing radiations and chemicals. Rad. Res. Supplement No. 1, 403–416.

Dale, W. M. 1953. The effects of X-rays on aqueous solutions of biologically active compounds. Brit. J. Radiology *16,* 171–174.

Dale, W. M., Davies, J. V., and Gilbert, C. W. 1949. Deamination of glycine by X-radiation from the disintegration of B in a nuclear reactor. Biochem. J. *45,* 543.

Dale, W. M., Davies, J. V., and Meridith, W. J. 1949. Further observations on the protection effects in radiation chemistry. Brit. J. Cancer 3, 31–32.

Dale, W. M., Davies, J. V., and Gilbert, C. W. 1949. Kinetics and specificities of deamination of nitrogenous compounds by X-radiation. Biochem. J. *45,* 93–99.

Desrosier, N. W. 1959. The Technology of Food Preservation. Avi Publ. Co., Westport, Conn.

Doty, P. 1959. Configurations of biologically important macromolecules in solution. Rev. Mod. Phys. *31*, 107–117.

Drake, M. P., and Giffee, J. W. 1957. Action of ionizing radiation on amino acids, proteins and nucleo proteins. Radiation Preservation of Foods. U.S. Govt. Printing Office, Washington, D. C. 133–158.

Drake, M. P., Giffee, J. W., Johnson, D. A., and Koenig, E. L. 1957. Chemical effects of ionizing radiation on protein. I. Effect of α radiation on the amino acid content of insulin. J. Am. Chem. Soc. *79*, 1395.

Errea, M. 1957. In Handbook of Protoplasm Research (in German) Springer. Vienna, Austria. 1–241.

Flory, P. J. 1953. Principles of Polymer Chemistry. Cornell University Press, Ithaca, New York.

Gordon, S. A. 1956. Auxin biosynthesis—a cytoplasmic locus of radiation damage. In Progress in Radiobiology. Oliver and Boyd. London, England. 44–47.

Gordy, W. 1959. Electron spin resonance of free radicals in irradiated biochemicals. Rad. Res. Supplement No. 1. 491–510.

Gray, L. H. 1959. Cellular radiobiology. Rad. Research Supplement No. 1. pp. 73–101.

Hollaender, A. 1954. Radiation Biology. McGraw-Hill Book Co., New York, N. Y.

Kauzmann, W. 1954. Denaturation of Proteins and Enzymes. In McElroy, W. O., and Glass, B. The Mechanisms of Enzyme Action. The Johns Hopkins Press, Baltimore, Md., 70–110.

Lea, D. E. 1956. Actions of Radiations on Living Cells. Second Ed. Cambridge University Press, Cambridge, England.

Leventhal, C. 1959. Genetics and the role of nucleic acids. Rev. Mod. Phys. *31*, 227–232.

Long, L., Jr., and Lirot, S. J. 1957. Action of ionizing radiation on carbohydrates and polysaccharides. Radiation Preservation of Foods. U. S. Govt. Printing Office, Washington, D. C. 126–132.

Lumry, R., and Eyring, H. 1954. Conformation changes of proteins. J. Phys. Chem. *58*, 110–120.

McArdle, F. J., and Desrosier, N. W. 1955. Influence of ionizing radiations upon some protein components of selected foods. Food Technol. *9*, 527–532.

McElroy, W. D., and Glass, B. 1959. The Chemical Basis of Heredity. The Johns Hopkins Press, Baltimore, Md.

Neuberger, A. 1958. Symposium on Protein Structure. John Wiley and Sons, Inc., New York, N. Y.

Neurath, H. 1959. Protein structure and enzyme action. Rev. Mod. Phys. *31*, 185–190.

Neurath, H., Greenstein, J. P., Putman, F. W., and Erickson, J. O. 1944. On protein denaturation. Chem. Rev. *34*, 157.

Pigman, W. 1957. The Carbohydrates. Academic Press, Inc., New York, N. Y.

Platzman, R. L., and Franck, J. 1958. In Symposium on Information Theory in Biology. pp. 262–275. Pergamon Press, New York, N. Y.

Pollard, E. 1959. Radiation inactivation of enzymes, nucleic acids and phage particles. Rev. Mod. Phys. *31*, 273–281.

Rich, A. 1959. Molecular structure of nucleic acids. Rev. Mod. Phys. *31*, 191–199.

Sanger, F. 1956. Currents in Biochemical Research. Green, D. E., Ed. Interscience Publishers, Inc., New York, N. Y.

SCHWEIGERT, B. S., and DOTY, D. M. 1958. Chemistry of color, flavor and odor changes in irradiated meats. Research Dept. S-507. U. S. Army QM. Food and container Institute, Chicago, Ill.

SIU, R. G. H. 1957. Action of ionizing radiations on enzymes. Radiation Preservation of Food. U.S. Govt. Printing Office, Washington, D. C. 169–176.

SMITH, C. L. 1953. The inactivation of deoxyribonuclease by electron bombardment, deuteron bombardment, and heat. Arch. Biochem. and Biophysics *46*, 83–90.

STOCKEN, L. A. 1959. Some observations on the biochemical effects of X-radiation. Rad. Research Supplement No. 1, 53–72.

SWANSON, M. A. 1948. Degradation of polysaccharides by enzymes. J. Biol. Chem. *172*, 805–814.

SWANSON, M. A., and CORI, C. F. 1948. Structure of polysaccharides. I. Acid hydrolysis of starchlike polysaccharides. J. Biol. Chem. *172*, 792, 797–804.

WHISTLER, R. L., and SMART, C. L. 1953. Polysaccharide Chemistry. Academic Press, Inc., New York, N. Y.

WOLFF, S. 1959. Interpretation of induced chromosome breakage and rejoining. Rad. Research Supplement No. 1, 453–462.

YOCKEY, H. P. 1958. Symposium on Information Theory in Biology. Pergamon Press, New York, N. Y.

CHAPTER 11

The Living Cell

The Basic Unit

Protoplasm is the only form of matter in which the phenomena of life are manifested. The concept that all plants and animals are composed of fundamental units called cells was developed in 1839 by two German scientists, Schleiden, a botanist, and Schwann, a zoologist.

Most cells are too small to be visible; micro-organisms numbering in the hundreds of thousands may conveniently be arranged on the period at the end of this sentence. Some nerve cells in animals and fiber cells in plants extend for a yard, and a quarter in U. S. A. currency placed on the leaf of a plant would probably cover about a million cells. Living cells vary in size from the most minute virus (10^{5} mm.) to such giant cells as eggs, and all exhibit the phenomena of life.

Many cells are smooth and regular, exhibiting a remarkable economy of exposed surface. Other cells are irregular, some being long and thin while still others exhibit many apparently unnecessary abberations from the regular simplicity of shape which we might expect. Cells may be either rugged individualists or "organization" cells, but all are fundamentally very similar. They have a boundary called the cell membrane, a body called the cytoplasm, and a third common structure—the nucleus.

The irradiation of single celled organisms, leads to a variety of detectable effects, e.g., altered nutrient requirements, changes in membrane permeability, changes in shape, gene mutations, failure to reproduce, and death. In higher animals the radiation effects express themselves in rather different form, such as deformed growth, radiation sickness, sterility, and death. These symptoms are gross manifestations of damage to one or more of the various specialized groups of cells; the irradiation of bone marrow for example may cause death to an entire vertebrate organism.

In the end the effect of irradiation on any living organism should be deducible from the effects on one or more particular cell types and their components. And, ideally, the end results of the radiation treatment should be deducible also from the precise role which these particular cells play on the stage of life. However, there is not sufficient information presently available to permit this deduction except in isolated cases.

Man lives in constant association with an environment in which living organisms composed of single cells play a not negligible role. These organisms are either helpful or harmful to his existence (for example, yeast cells in the fermentation industry or poisoning organisms in his food).

The effects of radiation on single-celled organisms offer possibilties of better control and utilization of these organisms by man. Furthermore, the study of single organisms gives an insight into phenomena of the interaction of radiation with living organizations. In this chapter the structure and dynamic processes of single cells will be reviewed and, in the next, the effects of irradiation on such cells as best is known will be evolved. The technological importance of these radiation effects will then be explored, followed by the problem of multicellular organisms in subsequent chapters.

The Structure of Cells

Living cells are enclosed in a membrane made from protoplasm, and this membrane is a living functional part of the cell, most important in regulating cell contents. The membrane is extremely thin (upwards of 60 Å). Experimental evidence concerning the nature of this membrane indicates it to be composed of a network of protein plates, with lipid molecules organized and oriented perpendicular to the surface (Frey-Wyssling 1957). Within the cell membrane the unspecialized protoplasm is called cytoplasm, Figs. 47 and 48. The cytoplasm contains many granules, some of which are recognized as centers of cellular metabolism, mitochondria, and microsomes. The mitochondria, present in all cells in the

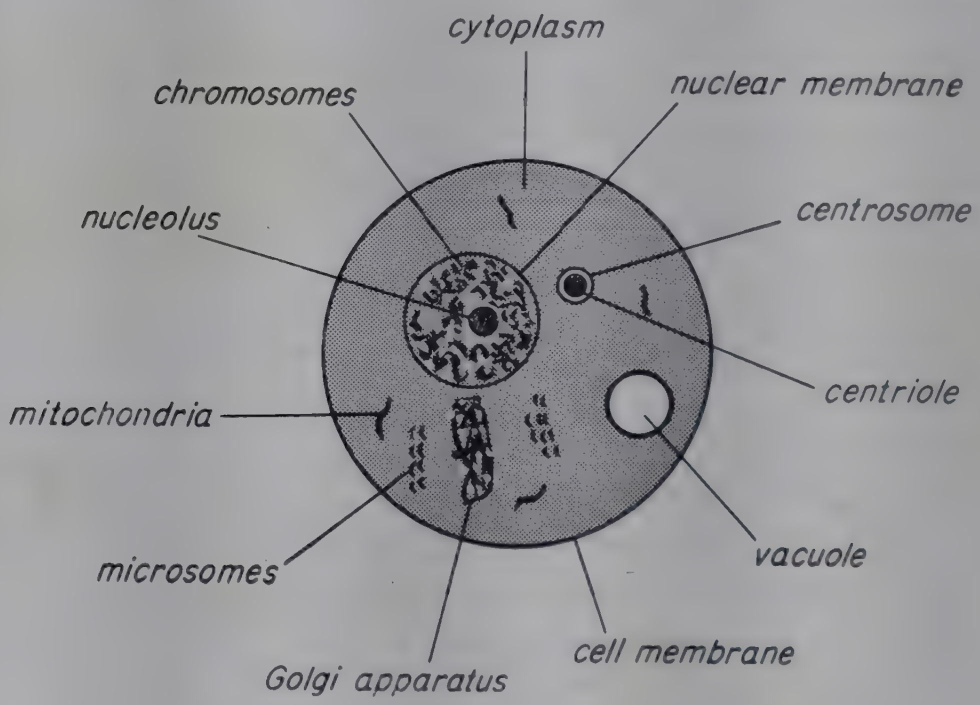


Fig. 47. Schematic Diagram of Animal Cell

cytoplasm, appear as sausage shaped objects, and are most numerous in the part of the cell with the highest metabolic rate. Microsomes are smaller but equally vital, both being composed of phospholipids and proteins.

Plant cells contain plastids which are larger than the other granules and are proteinaceous materials in the cytoplasms. One type of plastid is called a chloroplast which contains the pigment chlorophyll, significant in photosynthesis. The leukoplasts are another type which act as centers for the storage of materials in the cytoplasm. A third type, the chromoplasts, are the color centers. The mitochondria are believed to be involved, either directly or indirectly, in plastid formation; it has been noted that mitochondria may be found in clusters from which plastids appear.

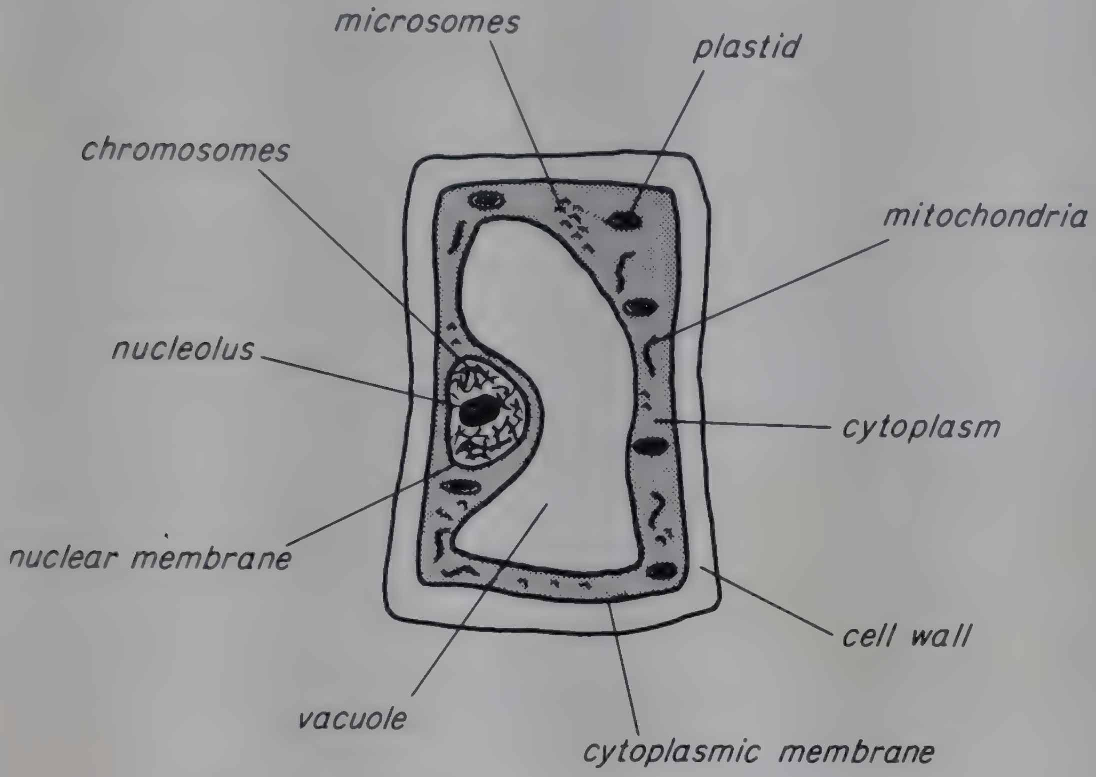


Fig. 48. Schematic Diagram of Plant Cell

In the cytoplasm of animals cells, located near the nucleus, is a small inclusion called the centriole. The centriole is active at the time of cell division, but is not found in plant cells.

Golgi apparati are also found in the cytoplasm, appearing as rods or canal type systems and distinguishable from mitochondria and plastids. Mitochondria are heavier than protoplasm while the Golgi apparatus is lighter. Plastids have a much different shape.

Each cell contains either a clearly defined nucleus or distributed nuclear material. In some cells the position of the nucleus is fixed centrally while in others it may appear in different positions within the cell. The nucleus is composed of a nuclear sap or nucleoplasm surrounded by the nuclear membrane which controls the flow of materials in and out of the nuclear structure. While the cytoplasm appears as a viscous translucent material with granular structures in suspension, the nuclear material is more dense and running irregularly through it are strands of chromatin, which at the time of cell division become recognized as the chromosomes or rod-shaped structures which bear the genetical machinery. The nucleolus is a rather spherical object within the nucleus, variable in structure, sometimes changing its form, appearing singly or in multiple units. The nucleolus is involved in cell division; it disappears during the process and reappears afterwards.

While the above components of cytoplasm are considered its living elements, cytoplasm may also contain vacuoles or spaces filled with a watery fluid and separated by a membrane from the cytoplasm. Vacuoles are commonly found in the cells of both plants and lower animals. The nutrients or food for the cell, not a part of the living protoplasm, are found within the vacuole system. A tabular listing of the structural components of typical plant and animal cells is given in Table 25.

There are then three large differences seen in the structure of plant and animal cells. Animal cells generally have a centriole. Plant cells usually have plastids in the cytoplasm. Plant cells have a stiff rigid cell wall of cellulose which restricts their change of position or shape, while animal cells usually have but a thin plasma membrane which is flexible and has no cellulose. The plasma membrane of both plants and animals are generally alike in other regards.

Although the structure of protoplasm is generally sub-microscopic, components of the life substance are highly organized into functional units which are identifiable. As a result of numerous investigations of protoplasm, a number of hypotheses have been developed regarding its physical organization as observed through the microscope. These hypotheses include the granular, alveolar, and reticular. The granular hypothesis suggests that protoplasm is composed of tiny granules suspended in a liquid medium. The alveolar hypothesis sees protoplasm as a system of overlapping globules. The reticular hypothesis suggests that protoplasm is a network structurally of interwoven fibers with the interstices being occupied by water containing various dissolved and suspended components. Granules and globules are observed in protoplasm, however, and cannot be disregarded as basic structures of the life substance. Current concepts are that long protein molecules are the structural units, being held together at points of contact by chemical bonds, probably hydrogen bonds. This view is consistent with the properties observed for protoplasm in general (Schmitt 1959). It would appear that globules, granules, and

Table 25

STRUCTURES OF LIVING CELLS

- Plant Cell
  - cell wall
    - middle lamella
    - primary wall
    - secondary wall
    - pores
  - protoplasm
    - cytoplasm
      - pores
      - undifferentiated cytoplasm
      - cytoplasm membrane
      - plastids
        - chloroplasts
        - chromoplasts
        - leucoplasts
      - mitochondria
      - microsomes
    - nucleus
      - nuclear membrane
      - nuclear sap
      - nucleolus
      - chromosomes (chromatin)
  - vacuole
    - fluid
      - water
      - solutes
      - colloidal materials
      - crystals
    - membrane
- Animal Cell
  - cell membrane
    - membrane
    - pores
  - protoplasm
    - cytoplasm
      - undifferentiated cytoplasm
      - mitochondria
      - Golgi apparatus
    - centrisome
      - centriole
      - centrosphere
    - nucleus
      - nuclear membrane
      - nucleolus
      - nuclear sap
      - chromosomes (chromatin)
  - vacuole
    - fluid
      - water
      - solutes
      - colloidal material
      - crystals
    - membrane

other bodies in the protoplasm may be a superstructure on a basic protein network crudely seen as the life substance.

Cell Structure in Relation to Function

In a recent discussion of molecular biology and the physical basis of the life processes, Schmitt (1959) has analyzed the significant discoveries. By fragmenting cells, it is possible to prepare fractions of various cellular components and study the biochemical and structural properties of these organelles.

It has been found that the mitochondria are the source or the power plant of a cell in which, through oxidative phosphorylation, high energy phosphate bonds yield energy through the splitting of adenosine triphosphate (ATP), which will be seen later as the fuel for cell metabolism. The striking aspect of mitochondrial organization is the internal layered structure. It also appears that while the mitochondria are of vital importance in metabolism, they are not the ultimate elementary unit (Weiss 1959).

Studies have shown that both the cytoplasm membrane in an animal or the internal lamella of a plant cell contain the enzymes required for the energy cycle to be discussed below. It is thought that the individual enzymes involved in the cycle occur anchored in or on the cell membranes (Lehninger 1959). With fragmented cells it has been possible to demonstrate spectacular biosynthetic production of not only peptides and steroids, but other complex molecules as well, when appropriate precursors are supplied to the fragmented cells (Hogness 1959). For example, simple protein has been synthesized from peptide fragments. Sufficient amounts of active cell surfaces are retained in such ruptured cells to accomplish biosynthesis, confirmed by using radioisotope tracer techniques.

Microsomes are lighter than the mitochondria and hence separable by differential centrifugation, and similar biosynthetic actions have been found (Schmitt 1959). However, the microsomes appear to be fragments of the membrane-limiting structure of the cell which when associated with ribonucleic acid (RNA)-rich particles (to be seen later to be manufactured in the nucleus) are currently thought to be the biosynthetic center of a cell.

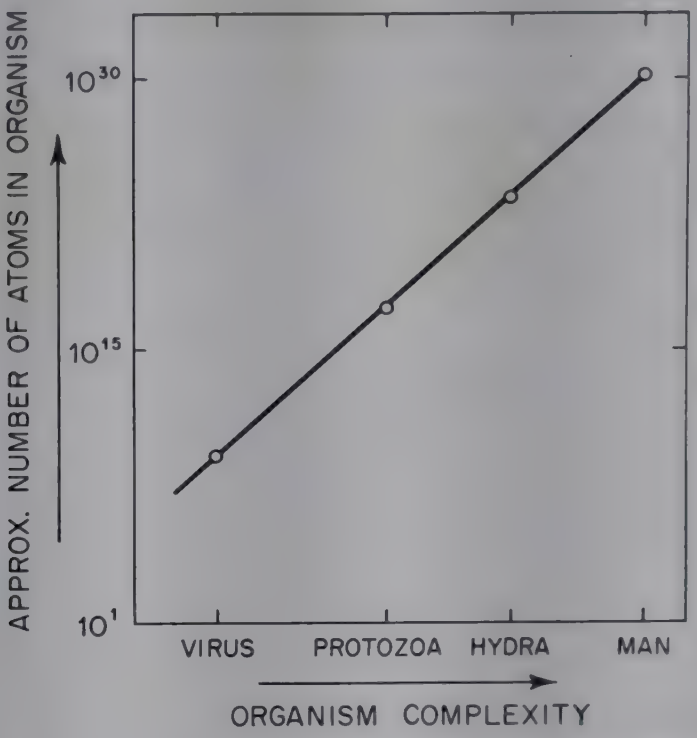
The cytoplasm appears to be separated into two phases. One phase is continuous between the nuclear membrane and with extracellular fluid through micropores in the limiting cell membrane. This material is fluid, homogeneous, and perhaps contains the biosynthesized components. A second phase, then, would consist of the mitochondria, the microsomes, and the RNA-rich granules which appear to be the actual network for biosynthesis. Membranes must be seen as metabolically very active surfaces, separating regions of the cytoplasm from one another (Berliner 1959).

The Golgi apparatus and the plasma or cell membrane are also of substantial importance. The Golgi apparatus appears to be a multi-layer canal-like structure in that section or zone of the cell which is actively synthesizing. It is thought that the Golgi apparatus is a membraneous structure which surrounds or encloses secretion granules or particles which by fusion to the cell membrane serve not only to contain the cell and to control the flow of molecules in and out, but also to assist the biochemical mechanisms by transporting components (Roberts 1959).

As the functions of the identifiable parts of the cell, such as the microsomes, mitochondria, Golgi apparatus, and limiting membranes become better understood, it is becoming increasingly apparent that the organization of a cell is most complicated and each artifact seen in a microscope has one or more specialized functions (Sjostrand 1959, Weiss 1959). Somehow the required reactions occur in the right locations at the proper time in a given cell or in the organism as a whole to achieve that complex pattern of life genetically transmitted.

During the past decade sufficient information has become available to form the basis of a new area of investigation called the chemical nature of inheritance (Mirsky 1953). These studies have been concerned with two main problems, one, the chemical and physical nature of chromosomes and their genes, and two, the mechanisms by which genes control the development and maintenance of an organism. It has been found that the genes chemically are nucleoproteins, which are composed of proteins and nucleic acids. The latter are found in all biological substances capable of life.

Rich (1959) has drawn the following analogy between the function of nucleic acids in cells and the function of the punched tape used in an electronic computer. Both punched tapes and nucleic acids are very long elements which direct a larger unit, either the computer or the cell in which they are located. Both the tape and the nucleic acids carry information through variation in linear arrangement of a few fundamental repeating units along their length. Nucleic acids are long polymeric molecules built up by the repeated connection of a very few small molecules. It is generally thought that the specificity of nucleic acid functions arises from the particular sequence of its constituent residues as well as from geometric configuration. It is further believed currently that the deoxyribonucleic acid (DNA) functions as the ultimate carrier of genetical information (Butler 1959). All the cells of a given organism have the same organized DNA content with very few exceptions, and in these exceptions the content appears in one-half the normal amount. The DNA in all tissues of a given animal are found to be the same, and it is thought that all cells of an organism contain the same set of DNA molecules. The DNA is located in the chromosomal material of the nucleus, and during cell division the DNA is replicated so that an equal amount is found in the daughter cells produced with the same chemical composition found in the parent cells. One of the great achievements will be the discovery of the fundamental nature of this replication process. Although the amount of DNA is the same in each cell of a given organism, the amount varies from species to species (Rich 1959). In general the more complex species have more DNA. In analogy, the instructions for the solution of more complex problems require that a greater amount of tape be threaded into the electronic computer. An interesting calculation by Butler (1959) is shown graphically in Fig. 49 relating the general complexity of organisms to the approximate numbers of atoms present. Complexity is used here in an intuitive sense rather than a quantitative one.



Courtesy of Butler

FIG. 49. RELATIVE COMPLEXITY OF ORGANISMS IN TERMS OF THE NUMBER OF ATOMS PRESENT

The subtle but definite interaction of radiation with such intricate systems is to be anticipated.

Dynamic State of a Cell

Weiss (1959) conceives of a cell as a system of highly complex, but ordered molecular populations grouped in a hierarchy of supra-molecular complexes in constant interaction among themselves and with their environment (the space beyond the cell border) leading to features the visible expressions of which are recorded as shape. When the environmental conditions are markedly changed, the cells of a group of the same type may vary from what might be considered normal. For example bacteria are sensitive to temperature effects. Acetic acid bacteria at 45°F. grow slowly and the cells are short and usually broad. At 70°F. the cells appear normal, growing and developing into chains of cells of varying numbers and cell walls become swollen. At 100°F. long thread-like transparent filaments with no visible cross walls and irregular bulgings with even some branching have been observed. Interestingly, lowering the temperature to 70°F. results in the production of some cells with "normal" characteristics and behavior.

METABOLISM

Recognizing that the chemical substances present as protoplasm are very complex, it is to be anticipated that the chemical reactions which occur in protoplasm may be correspondingly complicated. The sum of the chemical activities of protoplasm which provide for its growth, maintenance, and repair is called metabolism. Living protoplasm is a dynamic substance; nutrients are taken in, new protoplasm is built, and a part of the energy contained in large molecules such as carbohydrates, fats, and proteins is made available as these nutrients are changed to simpler compounds. This constant exchange of energy is unique and characteristic of living entities. The chemical reactions which are found characteristic of life are divided into two groups. Anabolism refers to those processes in which simple molecules are united to form complex materials. A part of the activity involves the storage of energy and the manufacture of new protoplasm and growth. On the other hand catabolism refers to the breakdown of the complex molecules resulting in the release of energy and to the use and destruction of protoplasm. Both types occur continuously and life is an intricate combination and balance of both. Death is an irreversible imbalance. Both plant and animal metabolism have anabolic and catabolic phases.

The fascinating performers in a living cell are the enzymes, the catalysts of life. As discussed in Chapter 10, an enzyme is similar to a catalyst but manufactured by the living organism; man, plants, and animals are in a sense elaborate enzyme machines. Enzymes despite their small quantity are essential as the machinery which apparently guides, achieves, and integrates the whole series of chemical reactions we call the life process As will be seen below, enzymes are controlled initially by the nucleus.

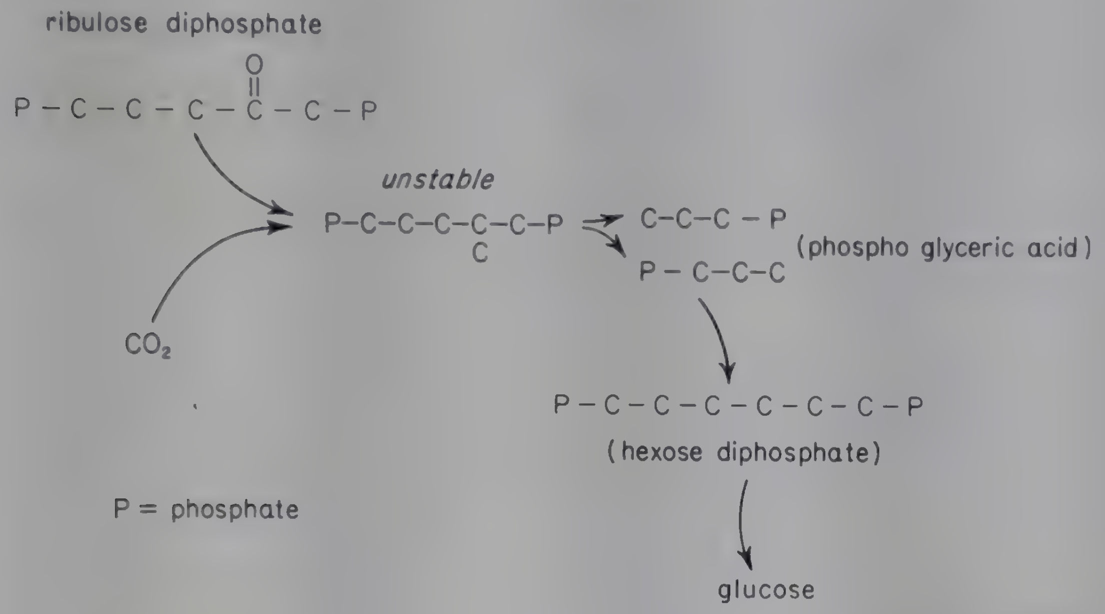
Energy for Cell Processes

Life is an energy demanding process; this energy, acquired from the sun is stored in the form of chemical compounds and released in a series of chemical reactions through the agency of enzymes. One facet of the life process is then seen as the utilization of energy for the elaboration of complex components recognized as the building blocks of living entities such as protoplasm. Cellular functions are driven by the energy obtained from the degradation of nutrient molecules. A cell requires energy to achieve biosynthesis, to do mechanical work, or to accumulate chemical substances internally.

Cells may be grouped into two large categories: those which obtain their chemical energy by aerobic oxidation of nutrients, molecular oxygen being required, and those which obtain their energy through internal oxidation-reductions by fermentation (rather than by respiration), which does not require oxygen. Respiration can be considered as the aerobic method of degradation of nutrients to obtain chemical energy and fermentation is the anaerobic method. The major energy accumulating cycle in nature is seen as the conversion of radiant energy from sunlight into chemical energy through the agency of photosynthesis. In this process, carbon dioxide is reduced to the oxidation level of carbohydrates. Respiration (or fermentation) and photosynthesis are then seen as two complementary accumulating and releasing processes for living cells.

Photosynthesis

The photosynthetic apparatus of a green plant is located in the chloroplasts which contain the pigment chlorophyll. Chlorophyll is capable of participating in photosynthesis only when in association with the components of chloroplasts. All chloroplasts show a laminated sheet-like structure, in multiples of two ultra thin (about 65 A) membranes, joined at the ends. A single membrane appears to consist of a film of protein with a carotenoid layer between it and the next layer (Frey-Wyssling 1957). The chlorophyll molecules are organized between the protein and carotenoid substances. The principal reaction carried on in the chloroplast is thought to be the splitting of water into two radicals, ·H and ·OH (Calvin 1959). Oxygen is eventually released from the ·OH; hydrogen atoms react with a suitable hydrogen acceptor. It is thought that a function of this film or membrane in the chloroplast is to keep the hydrogen and hydroxyl radicals apart and prevent their recombination. The critical process in photosynthesis is then the splitting of water into hydrogen and hydroxyl radicals and the subsequent use of the hydrogen atoms to bring about reducing reactions which result in the production of energy-rich compounds such as adenosine triphosphate (ATP). In the process carbon dioxide is taken from the atmosphere, an oxygen atom is removed, a hydrogen atom is added, converting the now reduced carbon dioxide into a fragment available to build into a carbohydrate. The photosynthetic carbon cycle which has been proposed by Calvin and associates (1959) is shown schematically in Fig. 50. As seen in the figure, carbon dioxide enters into a complicated cycle of changes. The sequence of rearrangements of hexose and triose are indicated through heptose and pentose, back again to ribulose diphosphate, which is then available to accept carbon dioxide and recycle.



Courtesy of Calvin

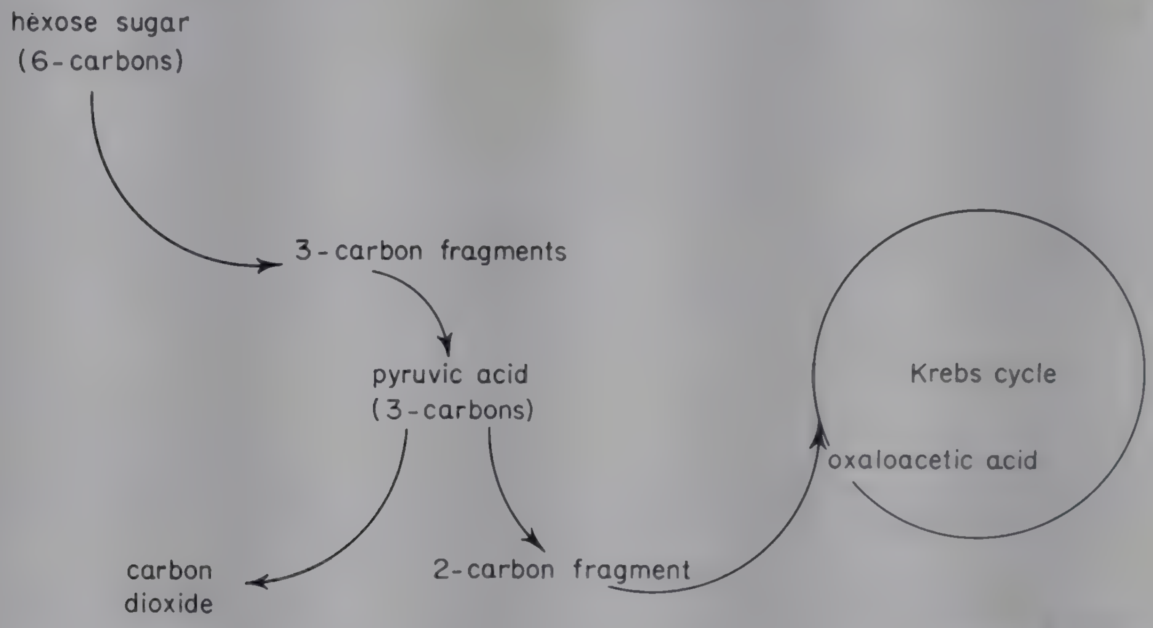
FIG. 50. SCHEMATIC DIAGRAM OF PATHWAY OF CARBON COMPOUNDS FROM CARBON DIOXIDE TO GLUCOSE IN PHOTOSYNTHESIS

A two second exposure of radioactive carbon-14 dioxide to the photosynthetic apparatus finds 90 per cent immediately recoverable in the three carbon "half" sugar fragment, thought to be the first stable component formed in photosynthesis (Calvin 1959). Longer exposures find carbon-14 in wide distribution in plant compounds ranging from carbohydrates to proteins to fats and vitamins and so on.

The formation of sugar and other carbohydrates is seen as not a simple, direct process but one which involves a large number of integrated stages. In the chapter dealing with the radiolysis of water, it was seen that ionizing radiation produces hydrogen and hydroxyl radicals from water. Butler (1959) has suggested a possibility that early in the history of the earth the process which led to living things used more penetrating solar (gamma) radiations for this synthesis. It might be noted at this point that Urey (1952) has suggested that high energy radiation might have been significant in the formation of protein precursors (amino acids) from inorganic matter in the era in which terrestrial life occurred. But in the photosynthetic mechanism of a living plant, a means has been found of using a low energy non-ionizing radiation, visible light, which is captured in the mechanisms of the chloroplasts. This chlorophyll system acts as an energy transformer, converting radiant into chemical energy. An amazing similarity of reaction is found in the mechanism of vision in higher animals through the agency of the retinal rods of eyes in converting visible light to chemical energy (Butler 1959).

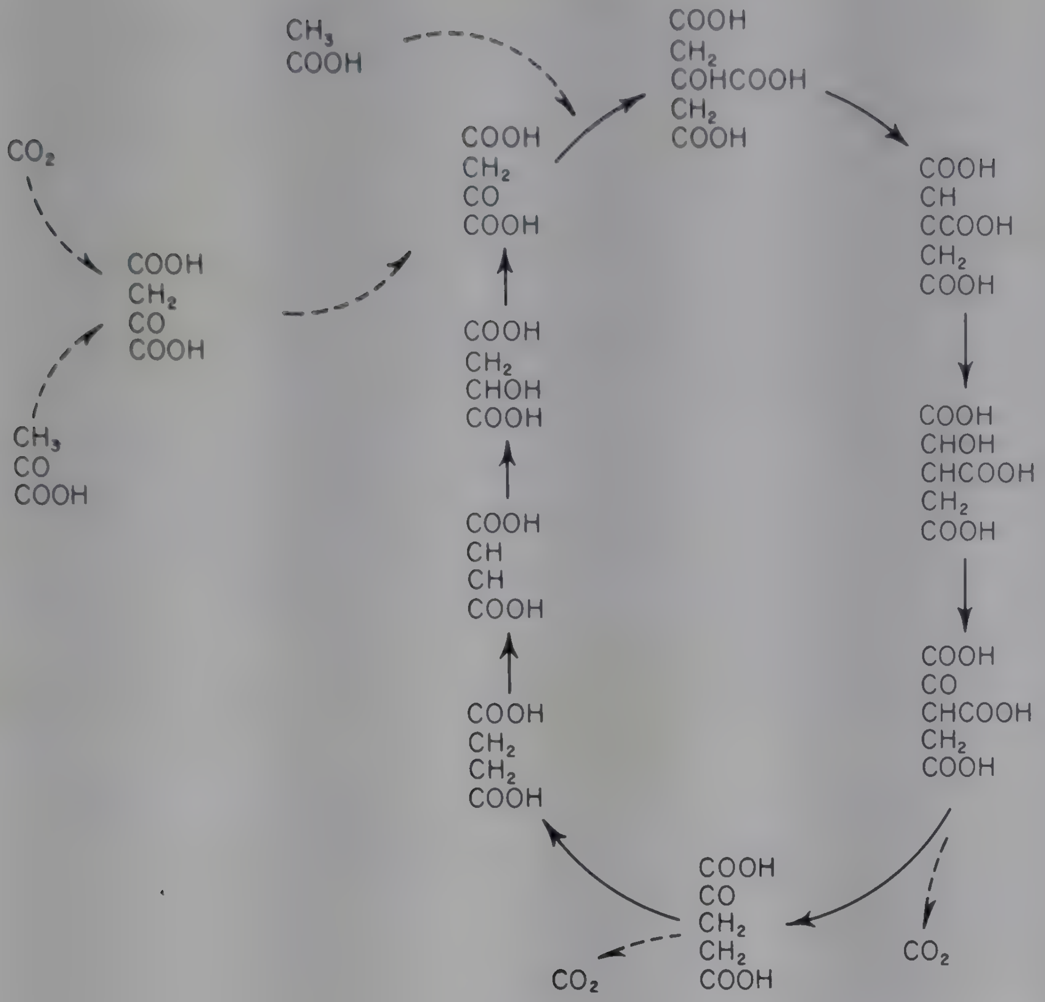
Respiration

While the photosynthetic process is unique in its yield of energy-rich chemical compounds, a cell expends energy in solely existing. The pathway for obtaining energy from carbon compounds in respiration is shown in Fig. 51. In simplified form, it is seen that a six-carbon sugar (glucose) is broken down into two three-carbon fragments. These are oxidized by removing two hydrogen atoms from each molecule in a reaction permitting the subsequent transfer of chemical energy from the carbohydrate to energy-rich phosphate (ATP). The reactions in the series include dehydrogenation and each step is mediated by an enzyme (see Figs. 52 to 54). This series of reactions is the main energy yielding process for cells by aerobic means (Roberts 1959). The manner in which this scheme operates is through a cyclic series of reactions known as the Krebs citric acid cycle. The net result is that pyruvic acid is oxidized to carbon



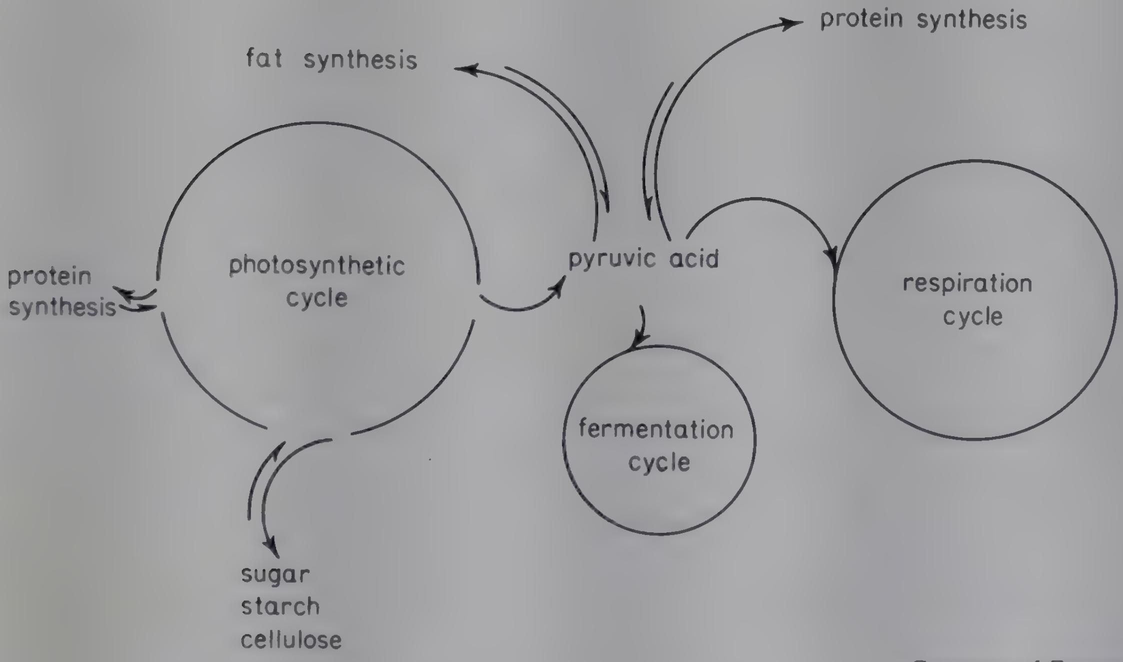
Courtesy of Calvin

FIG. 51. PATHWAY OF CARBON IN RESPIRATION


CH₃
COOH
CO₂
COOH
CH₂
CO
COOH
CH₃
CO
COOH
COOH
CH₂
CO
COOH
COOH
CH₂
COHCOOH
CH₂
COOH
COOH
CH
CCOOH
CH₂
COOH
COOH
CHOH
CHCOOH
CH₂
COOH
COOH
CO
CHCOOH
CH₂
COOH
CO₂
COOH
CO
CH₂
CH₂
COOH
CO₂
COOH
CH₂
CH₂
COOH
COOH
CH
CH
COOH
COOH
CH₂
CHOH
COOH

Courtesy of Roberts

Fig. 52. Flow Pattern of the Krebs Cycle for Cells Growing on Glucose as Carbon and Energy Cycle


fat synthesis
protein synthesis
protein synthesis
photosynthetic cycle
pyruvic acid
respiration cycle
fermentation cycle
sugar
starch
cellulose

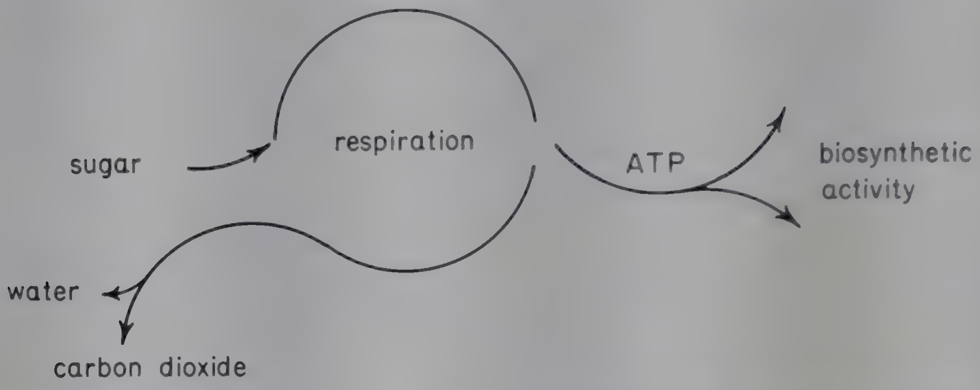
Courtesy of Bonner

Fig. 53. Metabolic Pathway in Plants

dioxide and water while the energy obtained in the process is stored as ATP, which goes into the pool available for all synthetic and work tasks of the organism or cells.

Transport Across Biological Membranes

In a single celled organism not only nutrient materials for the cell but also waste materials and other products given off by the cell during its life must pass a membrane system. The membrane is permeable to these substances but impermeable to others and therefore is said to be semipermeable or selectively permeable. In order to retain within a cell the required substances in the proper proportions and to prevent the entrance and allow escape of waste substances the cell membrane must exert a very fine discrimination at all times. Defects in cell membrane discriminatory properties must have a profound effect on cell contents and hence dynamics. The maintenance of the integrity and uniformity of the cell membrane is a matter of great importance. Substances or forces which tend to alter the wall or membrane or overcome its selective powers must influence the well-being of the cell.



Courtesy of and adapted from Bonner

FIG. 54. PHOSPHATE BOND ENERGY HARVESTED FROM RESPIRATION OF SUGAR

A number of possible ways are available by which materials may cross biological membranes (Berliner 1959). There may be a gradient of chemical activity including differences in concentration or activity coefficients. There are factors due to the gradient or electrical potential between surfaces. There are forces exerted upon solutes by the flow of the solvent through solvent filled channels or pores. In addition there are movements of solutes which are explained by none of these forces above, and which are designated as actively being transported, metabolic energy being utilized to perform the transportation effect. Naturally the manner in which solutes cross membranes depends upon the nature of the membrane. If it is a continuous lipid layer, fat soluble substances may pass by dissolving in it or a molecule may combine with some component of the lipid layer enhancing the solubility of the solute. There are features of cellular membranes which cannot be explained if they are assumed to consist of a solid lipid layer, however, and it is generally believed that this layer does in fact contain pores through which water forms a continuous phase and through which water soluble solutes pass. In other words, there are direct channels to cell cytoplasm. Berliner (1959), in reviewing membrane transport, finds that there is evidence that the solvent flow produces movements of solutes and that systems of solvent filled pores are present in membranes.

Cell membranes are composed of a lipid-protein structure penetrated by aqueous pores, the walls of which may bear an excess of fixed charged groups which will impede the passage of ionic species of the same sign. In multicellular organisms more complicated structures exist and may consist of an entire layer of cells attached to a common base membrane, in which case the passage of water soluble substances may also occur between cells (Bonner 1950).

One theory of ion transport through a membrane involves the redox potential. This theory is based on the assumption that oxidation and reduction of cytochrome iron inside the cell are especially separated and that hydrogen ions formed in the reductive step are available for exchange with some cation from outside the cell. Electrons are passed along the chain, the cations are taken up, possibly moving with the electrons.

It is found that the two surfaces which create a membrane, the inner and the outer, have somewhat different and specific properties. The two surfaces operate as diffusion potential boundaries specifically permeable to a single cation. This electrolyte transport by a membrane has received much attention. In the above cases, it appears that there is a net movement of water inward in an amount to make the net transport of the solute result in no net change in solute concentration in the cell. In the case of a pore permeating a cell membrane, impermeable to a solute on one side but not on the other, the effect of flow through such a pore accelerates the diffusion of molecules in the direction of flow and retards it in the opposite direction.

Cell walls vary in thickness, strength, and chemical composition, and also with the species and growing conditions and other unknown factors (Bennett 1959). Cells of the plant kingdom often have carbohydrate complexes such as chitin-like compounds, mucinoids, hemicellulose, and similar substances which are also porous, threaded through the lipid layer.

A schematic diagram of a membrane is shown in Fig. 55, indicating the orientation of lipid and protein components, enzyme prosthetic surfaces, cytoplasm channels, and water pores. It is to be recognized that the cell membrane is a living integral part of the cell and damages to this structure consequently damage the cell itself and its organization. If we view a cell as being a spherical mass, the outer surface of which is enclosed in a membrane, the inner surface of the membrane toward the cytoplasm being charged oppositely to the outer layer, it can be seen that the cell is in delicate balance between its internal components and the exterior environment with which it is intimately associated. The surface is punctuated with small pores which permit water molecules and certain solutes to traverse the membrane. In addition, active enzyme surfaces are located on the membrane surface, with the protein moiety residing in the cytoplasm. This forms a direct exposure and channel of cytoplasm itself with the outer environment of the intact cell. The cell membrane is therefore dynamic and capable of forming, sorting, and selectively absorbing and containing those components required for its biosynthetic life. In addition waste ingredients or components from metabolism must have a controlled avenue of exit. If one takes a position in a molecule on a living cell membrane surface, he would view a constant movement of molecules of varying complexity and number.

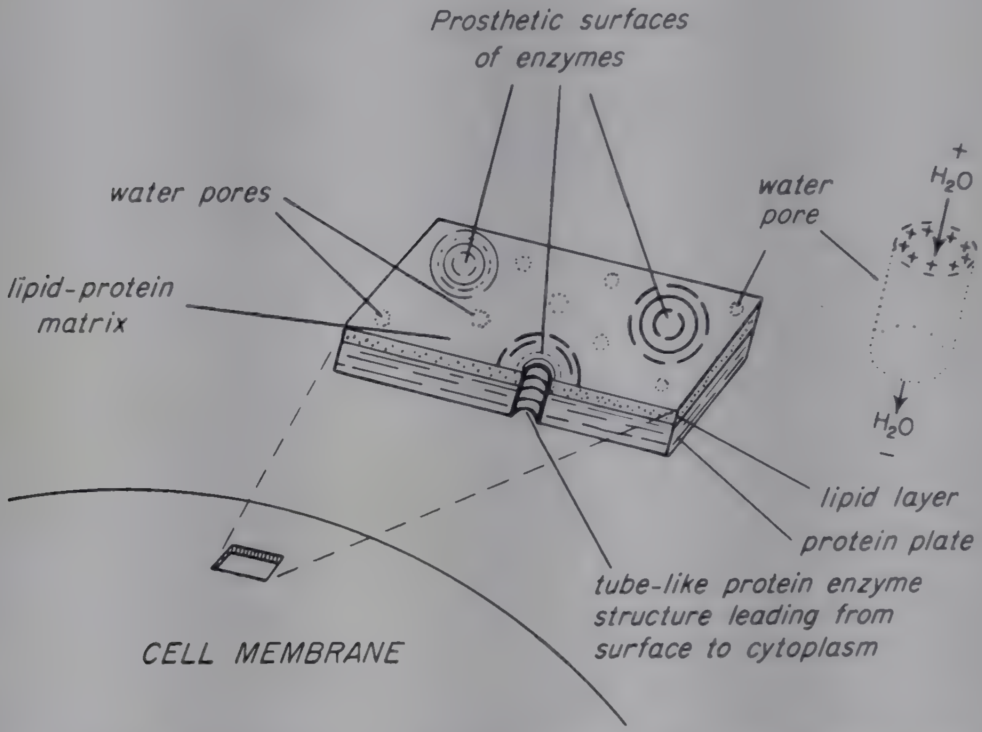

Prosthetic surfaces of enzymes
water pores
lipid-protein matrix
water pore
+
H_2O
H_2O
−
lipid layer
protein plate
tube-like protein enzyme structure leading from surface to cytoplasm
CELL MEMBRANE

Fig. 55. Schematic Representation of Cell Membrane Surface

The cell can do no better in its biosynthetic role than perform on the nutrients (chemical entities) available to it. Each species of organism has its own peculiar organization, arrangement and demands for nutrients.

and has its own particular assortment of enzymes. While throughout the plant and animal kingdoms there is an overriding similarity in metabolic mechanisms, there are occasionally some very subtle differences in the enzymes which are capable of performing similar tasks in different organisms. These are a part of the subtle but significant variations in various species, and perhaps represent alternate solutions to a common problem of equal value.

The problem of cell membrane nutrient transport is most complicated. Current thought is well reviewed in a variety of references. The striking similarities of the membrane transport phenomena in diverse systems leads investigators to believe that the processes involved, fundamentally, must be limited in number and basically not different in each cell type and species. The study of simple systems yields information applicable to the more complex and less accessible systems.

Aside from chemical interaction with radiation-induced free radicals, the effect of ionizing radiation on cells is the production of subtle changes in membrane structure. These changes may be brought about by the readjustment of the atomic arrangement of the membrane to the highly concentrated electrical charge distributions which are momentarily created by the passage of the ionizing particle. These structural changes have significant influences on the selectivity and permeability of the cell membrane. Some changes in nutrient requirements which may in fact be related to membrane permeability have been shown in single-celled organisms.

Nutrients and Nutrient Requirements

It has been said that the history of the science of nutrition has been one of finding that food consists of more than one nutrient and that these nutrients have specific physiological jobs. From about 1834 when the universal nutrient called food was found to be composed of three major components (carbohydrates, proteins, and fats) to the recent discovery of vitamin B_{12} (the animal protein factor) there have been more than 50 essential nutrients identified for one organism or another. The more complex nutrient requirement is associated with the more complex organism. The bacterium *Escherichia coli* requires but glucose and inorganic salts, while man requires the 50 odd specific nutrients. A listing of these chemical entities is shown in Table 26.

Organisms can be divided into major classes depending upon their requirements for an energy source. Autotrophs are chemosynthetic organisms using chemical oxidation, mainly of sulfur, ammonia, nitrites, or methane to obtain their energy. Their use of inorganic carbon is for synthesis and not as an energy source. The heterotrophs on the other

TABLE 26

CHEMICAL COMPOUNDS IN FOOD FOUND TO HAVE A ROLE IN LIVING ORGANISMS

Amino Acids:	*alanine, arginine, *aspartic acid, *beta-hydroxyglutamic acid, *cysteine, *cystine, *glutamic acid, histidine, *hydroxyproline, isoleucine, leucine, lysine, methionine, *norleucine, phenylalanine, *proline, *serine, threonine, tryptophane, *tyrosine, and valine.
Fats:	arachidonic, linoleic, and linolenic acids.
Carbohydrates:	sugar.
Minerals:	calcium, chlorine, cobalt, copper, fluorine, iodine, iron, magnesium, manganese, phosphorus, potassium, sodium, sulfur, and zinc.
Vitamins:	
fat soluble:	vitamin A (carotene), D (calciferol), E (alpha tocopherol), K (methyl, phytyl naphthoquinone)
water soluble:	vitamins of B complex, thiamin, riboflavin, niacin, biotin, choline, folic acid, inositol, pantothenic acid, para-amino benzoic acid, pyridoxine, B_{12} and vitamin C (ascorbic acid)

---

* Nonessential for man.

hand use organic carbon as a source of energy, a carbon compound as simple as ethyl alcohol being suitable for some species. Certain heterotrophs (viruses) are so highly specialized that they require their nutrients to be living protoplasm, being unable to grow in its absence. Some autotrophs are primitive in comparison, living solely on sulfur and a few inorganic salts in the presence of water. The general nutrient requirements of selected living organisms are shown in Table 27.

All living cells require inorganic elements which find specialized uses as parts of enzymes (copper and iron), in energy rich chemical bonds (phosphate ions), in maintaining osmotic pressure conditions in cells (sodium and potassium) and so on. Living cells are complex organic compounds of carbon, hydrogen, oxygen, and nitrogen with associated small quantities of inorganic elements as essential components.

TABLE 27

NUTRIENT REQUIREMENTS OF LIVING CELLS

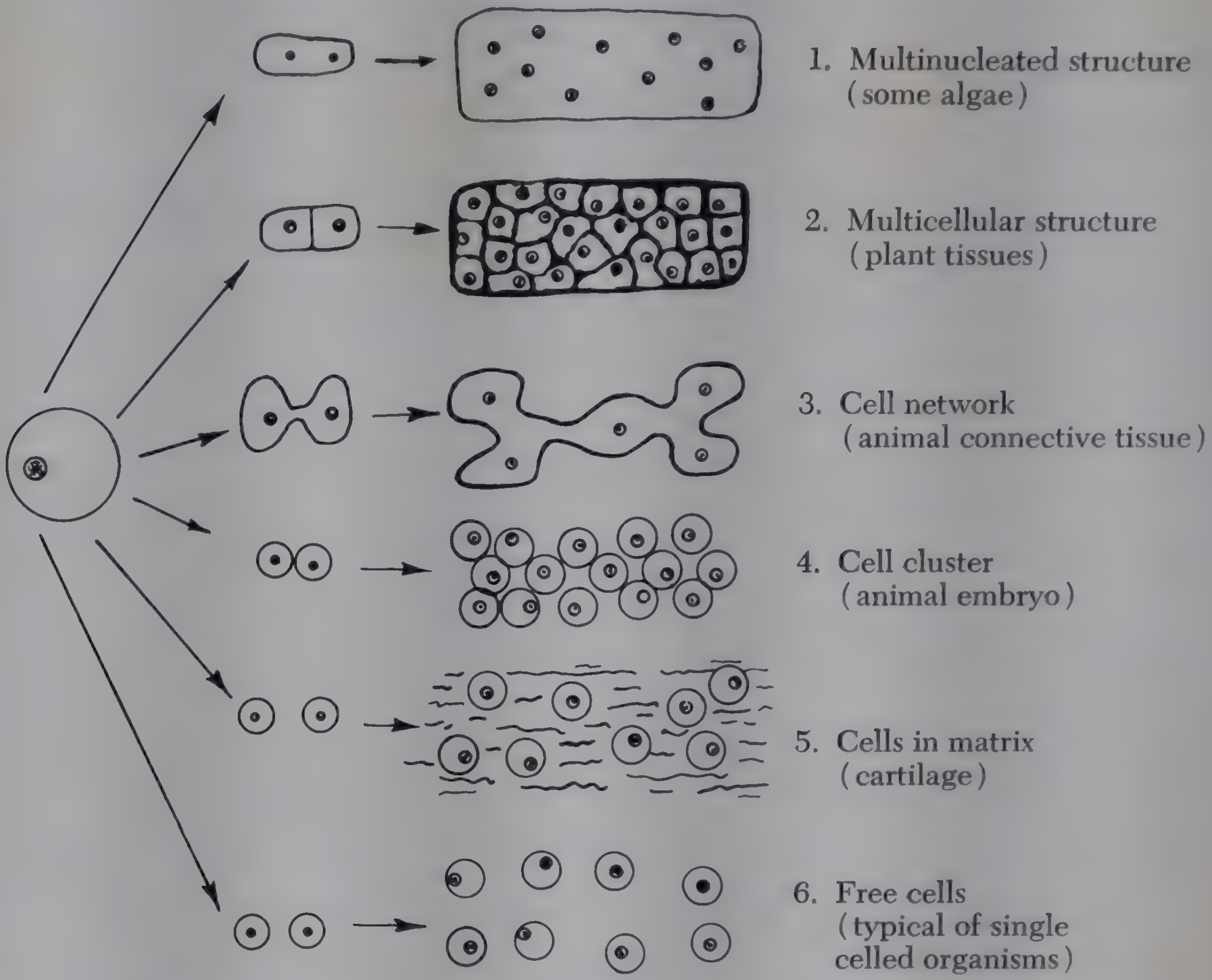
Group	Example
Soil autotrophs (nitrogen fixing bacteria and sulfur bacteria)	Require only inorganic salts and non-solar energy
Photosynthetic autotrophs (rhodobacteris, algae, and plants)	Require only inorganic salts and solar energy
Saprophytes and parasites (E. coli and Azotobacteria)	Use inorganic nitrogen and salts but organic carbon compounds
Higher saprophytes and parasites (micrococci)	Require organic nitrogen and carbon compounds and inorganic salts
Highly adapted parasites (diphtheria bacilli)	Require organic nitrogen and carbon compounds including vitamins and inorganic salts
Animals	Require specific complex organic carbon and nitrogen compounds, vitamins, and inorganic salts

The general life chemistry of a simple micro-organism is not different in principle from that of the other organisms. They all require water, carbon, hydrogen, oxygen, nitrogen, phosphorus, sulfur, sodium potassium calcium, and iron. While the fact remains that living organisms generally require much the same elements, in rather similar proportions, the form in which a specific element must be provided may vary substantially. Green plants require only carbon dioxide, water, and simple inorganic elements in the presence of solar energy of visible wave lengths. Animals require complex organic molecules, being incapable of elementary synthesis.

Cell Growth

In the preceding sections, we have viewed the structure of cells, identified cellular components, and dwelt on selected metabolic processes. We have seen how the nutrient requirements for organisms vary, and considered the interplay between a cell as a dynamic structure and its environment.

Growth of a cell is seen as the synthesis of new protoplasm under the guidance of the protoplasm which is already present. As a cell increases in size, certain results are achieved. A typical cell is envisioned as being composed of a spherical mass of cytoplasm surrounding a more or less spherical nucleus, these two components being inclosed in the cytoplasm membrane and a nuclear membrane respectively. Synthesis of new protoplasm involves an interchange of materials between the cytoplasm and nucleus from which is thought to emanate the instructions on which biosynthesis proceeds. The influence of the nucleus is determined in a qualitative sense by its membrane through which it contacts the cytoplasm. As a cell and nucleus increase in volume due to growth, the membrane through which the interchanges must occur does not increase at the same rate. A point is reached beyond which the volume and surface are insufficient to allow further organized growth of the entire protoplasm. In effect, what occurs is that the nuclear surface present in the protoplasm decreases in respect to the volume of cytoplasm that was originally present. This difficulty is solved simply by cell division. Growth after cell division can then proceed up to the point where this critical ratio of volume to surface again is reached. There appears to be a limitation to the growth potential of a spherical cell both in terms of the surface of the nuclear component as well as the surface of the entire cell itself. There is also an interplay between the external environment and the surface of the cell itself available for interaction due to the newly synthesized protoplasm. Here again the interference is overcome by the division of the cell. In some organisms, for example algae, the cell may then become flattened or filamentous in shape, to increase the surface per unit volume. In multicellular organisms, much more elaborate structural features are available to the organism which permit greater interaction between the composite cells and their environment. As protoplasm continues to grow, the patterns which the cytoplasm and nuclei may assume vary widely in different organisms. Sharp (1943) has made a diagrammatic representation for such development, shown in Fig. 56.


1. Multinucleated structure (some algae)
2. Multicellular structure (plant tissues)
3. Cell network (animal connective tissue)
4. Cell cluster (animal embryo)
5. Cells in matrix (cartilage)
6. Free cells (typical of single celled organisms)

Courtesy of Sharp

FIG. 56. PROTOPLASM GROWTH PATTERNS

1. Cytoplasmic growth may occur with the multiplication of nuclei without the separation of the cytoplasm into separate cells. The result is a multinucleated mass commonly seen in algae and fungi.

2. The multiplication may be accompanied by the development of membranes which do not separate, resulting in a multicellular mass familiar in the tissues of plants and animals.

3. As nuclei develop, the cytoplasm may not subdivide completely, but gradually form a network, remaining continuous from one area of nucleus to another. Such patterns are found in animal connective tissues.

4. After division the resulting cells may remain in a cell cluster, characteristic in animal embryos and also in the colony type algae.

5. After cell division, the individual cells which have been formed may become separated but accumulated in a matrix which has been secreted by the protoplasm during cell division and growth, commonly seen in cartilage.

6. With the formation of new cells they may become entirely independent and free as is the case in single-celled organisms such as bacteria. (With bacteria, however, no clear nuclear separation is identifiable.)

Differentiation of cells may then occur in any of the protoplasmic growths indicated above. The differentiation is not limited to multicellular organisms, as it is also seen in the unicellular protozoa which have rather clear digestive, excretory, and locomotion components, yet remain a single cell.

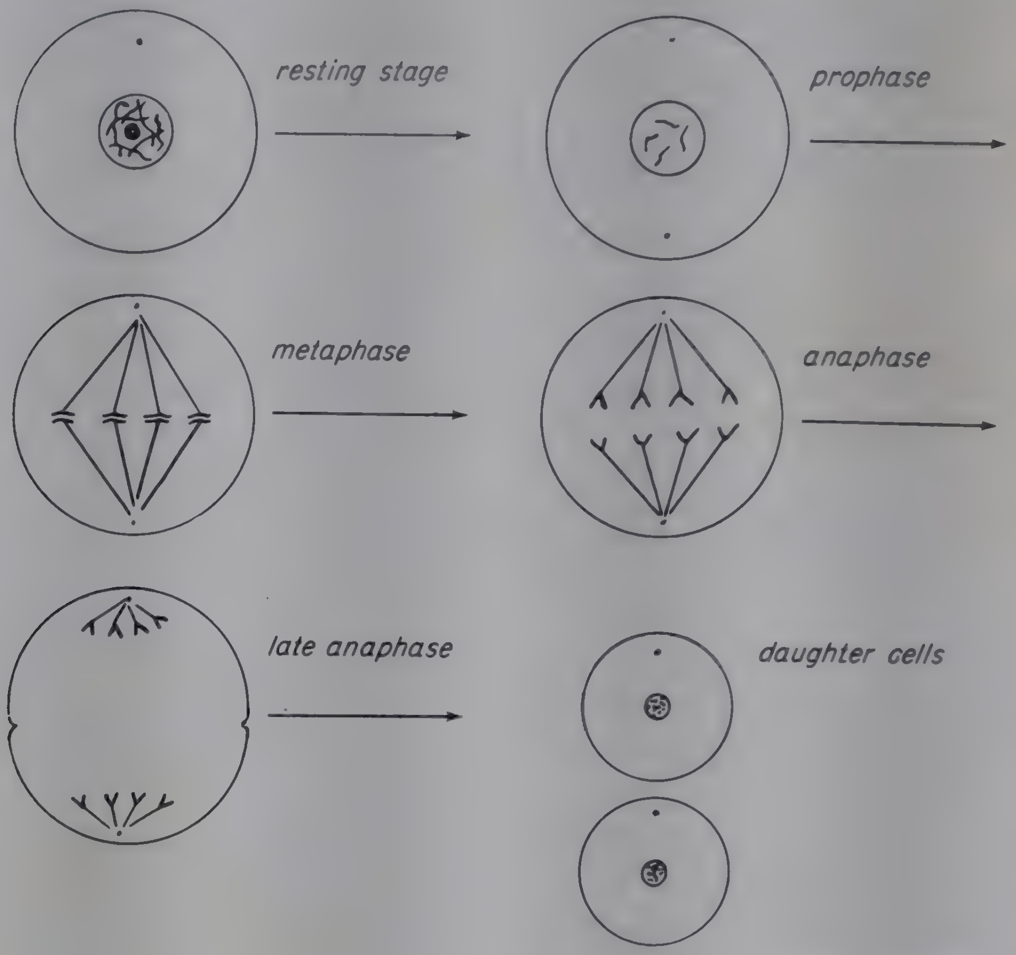
CELL DIVISION

Genes, located on chromosomes, exert their influence through enzymes and data from micro-organisms suggests that each gene has an effect on a particular enzyme. The question arises are the genes actually the manufacturing agents for enzymes. It appears to be reasonable that there are several steps between gene and enzyme and a number of intermediate substances are involved. Waddington (1953) has reported that both DNA and RNA are always present in those parts of a cell actively producing new substances and nucleic acid is essential for the production of any protein, and all enzymes are proteinaceous. RNA always occurs in high concentrations in regions of the cytoplasm where protein synthesis is proceeding rapidly. One theory proposes that DNA-containing chromosomes in the nucleus manufacture RNA which then passes into the cytoplasm and there becomes attached to the microsomes and take part in the synthesis of cellular proteins and enzymes. It is to be recalled that enzymes are the machinery which operate a cell and are responsible for integrating the process which is established. The process which appears to be established under the control of the genetical component is seen to play an active role in the actual synthesis of the enzymes themselves. The role of chemical messengers, including hormones, important in a harmonious functioning of cell communities such as higher animals, will be discussed in a later chapter.

The fundamental process of cell development includes growth and differentiation and it may take many patterns.

Mitosis

We turn now to the mechanism of cell division, the structure of the hereditary material, and the information which it carries. Mitosis is the process through which units of living matter duplicate themselves. The ability of cells to continue to grow in size is seen to be limited by geometric factors. Usually a cell can only double its mass and remain effective. As indicated above, when that condition arises in which surface to volume ratios are unfavorable, any further significant growth requires a production of more membrane surface, which is accomplished through cell division. Of the multitude of activities recognized to be occurring within a cell, mitosis is certainly the most spectacular and complicated process undertaken. Mazia describes the process as one of beautifully geometric perfection temporarily created out of the seemingly formlessness of a cell and operated with exquisite coordination. The sequence of events is shown schematically in Fig. 57. The process begins with a fully grown cell which has reached the critical surface to volume condition. At this stage there is no evidence of mitotic apparatus as it will develop. The component which is visible in animal cells is the centrosphere. The events appear as follows. The centrioles in the centrosphere separate, and push outward radiating fibers which are called asters. The asters



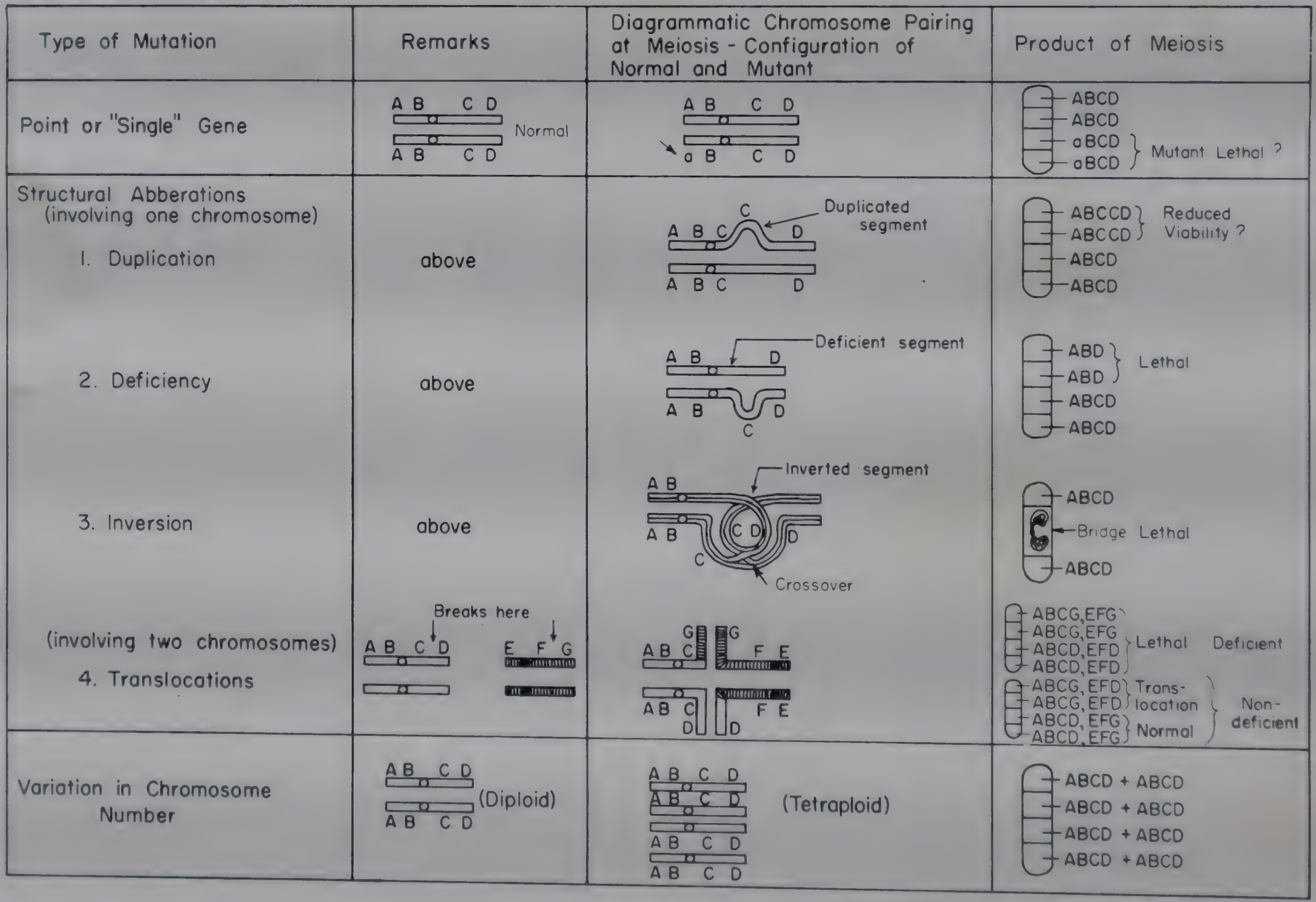
Courtesy of Mazia

FIG. 57. SCHEMATIC DIAGRAM OF MITOSIS

radiate from the two distinct centrioles as they move to opposite poles of the cell. In the meantime, the reorganization of the nuclear material has occurred. The seemingly disorganized nuclear mass begins to appear as distinct strands of definite but variable lengths. The asters which now connect one centriole to the other across the equator of the cell are called the spindle. It is thought that the chromatin material in the nucleus prior to cell division was an uncoiled form. With the beginning of the mitotic process, apparently, the chromatin material regains its tightly coiled form, and is then seen as the chromosomes. The chromosomes then become positioned at the equator of the cell, and exactly between the two centrioles and at right angles to the line connecting them. The apparatus is now organized for the cells to divide. At this point, each of the chromosomes has now duplicated itself, the duplication process occurring prior to the visible appearance of the two threads. In fact, it is thought that the duplication process occurs at the end of the resting stage which perhaps indicates that mitosis is a process for distributing the products of a reproductive step that had already been completed. The next dynamic stage in mitosis is the gradual movement of a complete set of chromosomes to each pole and this is co-ordinated with the division of the cell itself in such a way that each set of daughter chromosomes goes to one of the daughter cells. The mitotic apparatus is in action; the chromosomes are moved apart and become oriented in position near each centriole. Meanwhile, new cell membrane appears at the equator, forming two daughter cells, which then become entities in themselves each with a complete set of chromosomes seemingly identical to those of the mother cell. The newborn cells each retain a portion of the original cell membrane structure, it will be noted. The "new" cell then proceeds into the growth sequences described above.

The process has variations particularly between plant and animal cells. Animal cells seem to divide by a pinching effect; plant cells apparently build a new wall. Furthermore, in plant cells the appearance of the centriole and asters does not occur. However, in both cases the barrier between the daughter cells is formed exactly where the equator of the spindle was located. Single celled organisms may have rather unusual means of mitosis not necessarily taking the form of a sphere dividing into two daughter spheres. Examples are seen in the reproduction of protozoa, where longitudinal separation occurs.

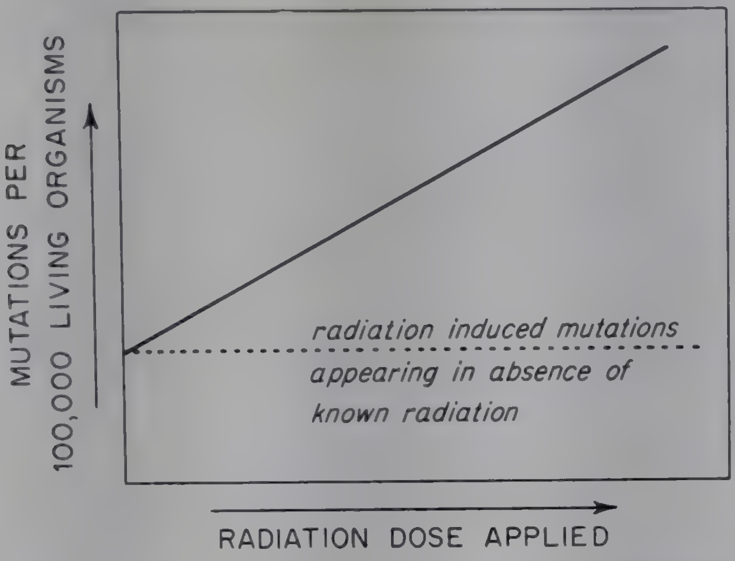
Mitosis can be a very rapid process; cells may pass through this entire cycle in less than a half hour. Preparation for the activity, however, may take more time than the actual division, which may occur in two or three minutes. In fact, some cells appear to snap apart once the pattern is set. Cell division is seen not as an increase in the amount of growing


Type of Mutation
Remarks
Diagrammatic Chromosome Pairing at Meiosis - Configuration of Normal and Mutant
Product of Meiosis
Point or "Single" Gene
A B C D
Normal
a B C D
ABCD
ABCD
aBCD
aBCD
Mutant Lethal ?
Structural Abberations (involving one chromosome)
1. Duplication
above
Duplicated segment
ABCCD
ABCCD
Reduced Viability ?
2. Deficiency
above
Deficient segment
ABD
Lethal
3. Inversion
above
Inverted segment
Crossover
Bridge Lethal
(involving two chromosomes)
4. Translocations
Breaks here
E F G
ABCG,EFG
ABCD,EFD
Lethal
Deficient
ABCG, EFD
Trans-location
ABCD, EFG
Normal
Non-deficient
Variation in Chromosome Number
(Diploid)
(Tetraploid)
ABCD + ABCD

Courtesy of Janick

FIG. 58. SCHEMATIC REPRESENTATION OF MUTATIONS

protoplasm of living matter; this only occurs during cell growth. Cell division permits continued growth. In higher organisms the pattern of development has been described as a counter-point of division and organization. About 50 successive divisions yield all the cells required by man. In an embryo the frequency of mitosis decreases before the forms and specialization of the adult come into being. When a group of cells reverses its pattern, the process is called cancer. When cells are exposed to ionizing radiation from which they recover, apparently what happens is that the synthesis of nucleic acid is suppressed. Increased doses of radiation further upset the mitotic process. Many biological manifestations resulting from an irradiation treatment involve the integrity of the chromosomes. Radiation effects on the mitotic system appear in interaction with the chromosomes (Figs. 58 and 59). A number of chemical compounds, for example colchicine, are also capable of interacting in mitosis.

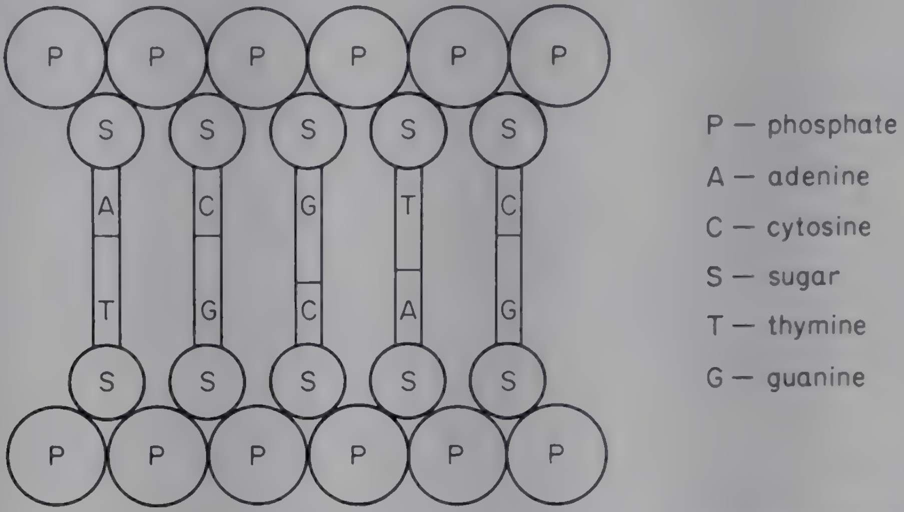


Courtesy of Bloom

FIG. 59. RADIATION INDUCED MUTATIONS

MECHANISM OF INHERITANCE OF CHARACTERS

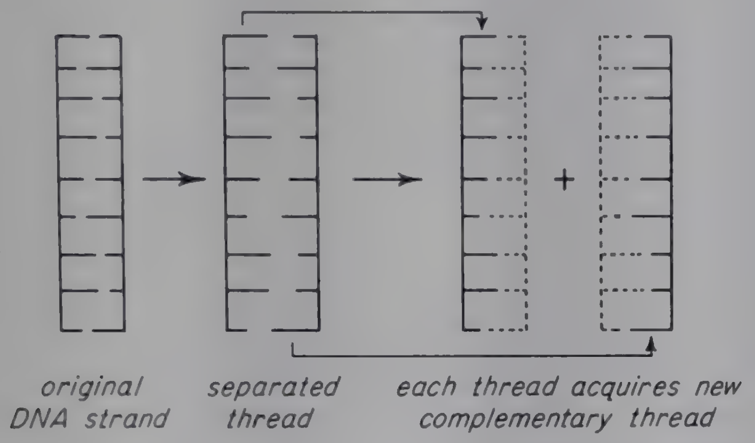
Chromosomes are intimately involved in the process of cell division. The original set of chromosomes is duplicated and each new cell has a full set of chromosomes except where mistakes occur, these mistakes being called mutations. Chromosomes are the carriers of the unit of hereditary characters called genes. Genes are believed to be arranged specifically in a linear sequence along the chromosome. As noted above, each gene appears to control one enzyme system. It is astounding that the chromosomes have oriented on themselves the genes, which are chemical compounds arranged in a pattern capable of maintaining their identity through many generations of cell divisions. They duplicate themselves in the process without mistake, or with very rare mistakes; the molecules of the genes control the processes whereby the new individuals develop, and thus perpetuate the organization. By subjecting these genetical components to chemical analysis it has been found that deoxyribonucleic acid (DNA) is the critical component; DNA is composed of sugar and phosphate groups occurring in equal number and arranged in long chains. There is also a protein moiety which is thought operative in binding the DNA into strands, the protein itself not being the information carrying agent, as it may vary in kind and amount, while the DNA is always present in a standard pattern for each species of organism.



Courtesy of Butler

FIG. 60. SCHEMATIC DIAGRAM OF CRICK-WATSON DOUBLE THREADED DNA

To each sugar molecule there is attached an organic base of one of four kinds, guanine (G), adenine (A), cytosine (C) and thymine (T). Strikingly, it has been found that the amount of adenine always equals the amount of thymine and the amount of guanine is always equal to the amount of thymine and the amount of guanine is always equal to the amount of cytosine. Watson and Crick (1953) have found that DNA is made up of two threads in which the adenine of one is paired with the thymine of the other, and the guanine is paired with cytosine in another thread. The schematic diagram of such a double threaded structure is shown in Fig. 60. Because their chemical structures are complimentary, this arrangement appears possible. Such a concept for the structure of DNA gives a clue to the possible mechanism for duplication or reconstruction of the threads. Each thread appears to be the exact complement of the other. If an A appears on one thread, a T will be found on the other and where a G is found on one, a C will be found on the other. Chromosomes are composed of large numbers of particles of DNA and it has been postulated that these particles are related to or actually are the independent genes. However, these are not the only components of chromosomes as some specialized proteins such as histones are also found. It may be that the histones are in some way protective of the DNA, although the mechanism is not known. When duplication of the chromosomes is undertaken, each new chromosome retains half of the old material and accepts half of the newly synthesized portion. The Crick-Watson model finds each DNA particle dividing into two simple threads and then each half reforming to complete the double thread schematically shown in Fig. 61. An important feature is that the DNA particles should maintain their position in linear order when duplication of the individual particles occurs. The fascinating feature is that each cell, when it divides, donates one gene of each kind to each of its two daughter cells and in the same order as they originally occurred. Occasionally, mistakes in reproduction occur, a gene may be out of order or may be omitted or may be by some accident reduplicated. These modifications are called mutations.



Adapted from Butler

FIG. 61. SCHEMATIC DIAGRAM OF DUPLICATION PROCESS FOR DNA PARTICLE. CRICK-WATSON MODEL

The whole picture which begins to emerge appears to indicate that genes, the hereditary particles, are composed of DNA, which is present in the cell nucleus and these are the information carrying or coordinating agents for the synthesis of specific proteins of a cell. The information appears first to be transferred to the RNA which then leaves the nucleus and enters the microsome granules (or the mitrochondria) where they form the template for the construction of protein molecules. It is thus seen that there is a mechanism for duplication of cell information centers which directs the synthesis of the enzyme systems required, which then achieve and integrate the series of events we recognize in the dynamic life process. The remarkable feature of genes in general is their permanence and ability to transmit a character through many generations without mistake. However, new characteristics do appear and old disappear. Butler (1959) reports work with the fruit fly in nature which on the average finds an individual gene undergoing a mutation in something on the order of a million duplications. This means that probably no mistake has been made in copying the original gene in perhaps a million attempts.

Haldane (1949) has calculated that with human genes, the likelihood of a useful mutation may be in the order of once in a million years. Nevertheless the fact that genetic mutations do occur is significant, permitting evolution to occur. It is to be noted that the majority of mutations are not advantageous and cause death to an organism. It is the rare occasion when a valuable mutation occurs in a population. The permanent nature of a gene and inherited characteristics led early investigators to conclude that there was no way of modifying genetical inheritance by external agencies. In 1928 Muller proved this to be incorrect, with his demonstration of the mutations possible using ionizing radiation (X-rays).

Chromosome Aberrations

Two features of the inheritance of characters are apparent. In the first place the genetical apparatus is one with great stability inherent in its design. On the other hand, evolution has occurred, and is occurring. A mutation is seen as a sudden variation in an off-spring which differentiates it from its parent in some well-marked character (see Fig. 60). Mutations may be either single point (single gene) phenomena or may involve more elaborate rearrangement, the latter occurring on a single chromosome or involving two chromosomes or whole sets. With single chromosome aberrations, three types have been encountered. If a normal chromosome is considered to have in linear arrangement genes 1, 2, 3, 4, and 5, to use a small number for simplicity, the three types of chromosome aberrations are seen to be gene inversion, duplication, and deficiency. Inversion occurs when the sequence of genes in the chromosome would be arranged 1, 3, 2, 4, 5. Duplication occurs when the gene arrangement appears 1, 2, 2, 3, 4, 5. A deficiency condition is found in which the gene arrangements may be shown to be 1, 2, 4, 5. A reciprocal exchange between two chromosomes is called a translocation. Strikingly, when one chromosome of a pair contains an inversion and passes through the meiotic process (when chromosome reduction occurs), a loop on one chromosome is formed during the pairing which allows the chromosomes to align, gene for gene. As a consequence of certain types of chromosome exchanges within the loop, two centromeres mav become attached to one chromosome, and a portion of the chromosome may appear without any centromere. (The centromere is seen as the fulcrum of a chromosome and is the entity which becomes attached to the spindle during mitosis and is drawn toward the pole, pulling the chromosome along with it.) The centromeres move to opposite poles, but in this case, remain connected by a "chromosome bridge." The chromosome fragment without benefit of a centromere becomes lost. These inversions result generally in lethal mutations due to the formation of deficiencies in a portion of daughter cells. Translocation may also result in deficiencies and lethality. The above chromosome aberrations are natural occurrences, and it is to be restated that most mutations are not useful, but in fact, are deleterious.

Point mutations involve changes within the gene. But what is the functional gene? The classic concept of the gene developed from 1900 to 1920 by Morgan (1928) and his coworkers considered genes analogous to beads on a string. Each "pearl" was considered a gene, capable of reproducing itself and manufacturing some substances in addition. The genes could exchange positions, as two strands of beads can be broken and recombined. The gene as a bead would not be affected. They could mutate; a bead could either be altered or change its position in the strand, and not affect neighboring genes. The unit of function, recombination, and mutation were considered to be the same bead. Recent information gathered during the 1930's and 1940's by many workers studying the genetics of the Drosophila fly and the corn plant indicated this bead concept to be inadequate.

The concept of a gene as a bead that mutates and recombines and is a physiological functional entity is no longer held. The work by Benzer (1957) can be cited as a case in point. The physiological unit of a gene itself can be identified and has been called the cistron. This is the smallest unit of a gene entity that will not function when further sub-divided. Furthermore, sub-gene particles or components are capable of recombining, and this smallest sub-division has been called the recon. Furthermore, the gene is composed of a finer structure, the smallest increment capable of evidencing mutation, and this is called the muton. Benzer (1957) was able to map such active areas along a single gene of a single virus.

Apparently, the functional genetic unit in the virus TII is composed of about 4,000 nucleotides. If the functional units average this length, the total DNA present in an average virus would provide for about 50 such units, assuming all the DNA present to be genetically active. While these 4,000 odd nucleotides apparently compose a single genetic unit, it now appears that these strings of nucleotides are arranged in a fine sub-structure recognizable in terms of the recon, the cistron, and the muton.

It is seen from the above discussion that even the smallest living cell is actually one of great complexity which the mind finds difficulty in grasping. Just how all these complex chemical components are arranged and fragmented and reassociated in particles and granules and oriented in protoplasm and how they achieve their functions in harmony with one another, producing what we recognize as life is hardly known. Some of the facets are becoming clear.

BIBLIOGRAPHY

ALBERTY, R. A. 1959. Mechanisms of enzyme action. Rev. Mod. Phys. *31*, No. 1, 177–185.

BENNETT, H. S. 1959. Fine structure of cell nucleus, chromosomes, nucleoli, and membrane. Rev. Mod. Phys. *31*, 297–301.

BENZER, S. 1957. The Chemical Basis of Heredity. W. D. McElroy, Ed. The Johns Hopkins University Press, Baltimore, Md.

BERLINER, R. W. 1959. Membrane transport. Rev. Mod. Phys. *31*, No. 2, 342–349.

BLOOM, W. 1959. Cellular responses. Rev. Mod. Phys. *31*, No. 1, 21–30.

BLUM, H. 1955. Time's Arrow and Evolution, Second Ed. Princeton University Press, Princeton, N. J.

BONNER, J. 1950. Plant Biochemistry. Academic Press., Inc., New York, N. Y.

BUTLER, J. A. V. 1959. Inside The Living Cell. Basic Books, Inc., New York, N. Y.

CALVIN, M. 1959. Energy reception and transfer in photosynthesis. Rev. Mod. Phys. *31*, No. 1, 147–162.

ELSASSER, W. 1958. The Physical Foundation of Biology. Pergamon Press, New York, N. Y.

FREY-WYSSLING, A. 1957. Macromolecules In Cell Structure. Harvard University Press, Cambridge, Mass.

HALDANE, J. B. S. 1949. The rate of mutation of human genes. Proc. 8th Intern. Genetics Congress. Stockholm, 1948. (Hereditas Suppl.) 267–273.

HOGNESS, D. S. 1959. Induced enzyme synthesis. Rev. Mod. Phys. *31*, No. 1, 256–268.

HOROWITZ, N. H. 1956. The Gene. Monogram 17, Scientific American, 11 pp.

JOHNSON, F. H., EYRING, H., and POLISSAR, M. 1954. The Kinetic Basis of Molecular Biology. John Wiley and Sons, New York, N. Y.

KAUZMANN, W. 1959. Chemical specificity in biological systems. Rev. Mod. Phys. *31*, No. 2, 549–557.

LEHNINGER, A. L. 1959. Respiratory-energy transformation. Rev. Mod. Phys. *31*, No. 1, 136–147.

MAZIA, D. 1953. Cell Division. Monogram 27. Scientific American, 11 pp.

MEYER, B. S., and ANDERSON, D. B. 1952. Plant Physiology. D. Van Nostrand Company, Inc., New York, N. Y.

MIRSKY, A. E. 1953. The Chemistry of Heredity. Monogram 28. Scientific American, 10 pp.

Morgan, T. H. 1928. Theory of the gene. Yale University Press, New Haven, Conn.

Müller, H. J. 1955. Radiation and Human Mutation. Monogram 29. Scientific American, 10 pp.

Rich, A. 1959. Molecular structure of the nucleic acids. Rev. Mod. Phys. *31*, No. 1, 191–200.

Roberts, R. B. 1959. General patterns of biochemical synthesis. Rev. Mod. Phys. *31*, No. 1, 170–177.

Schmitt, F. O. 1959. Molecular biology and the physical basis of life processes. Rev. Mod. Phys. *31*, No. 1, 5–11.

Schroedinger, E. 1945. What Is Life? Cambridge University Press, Cambridge, England.

Sharp, L. W. 1943. Fundamentals of Cytology. McGraw-Hill Book Co., New York.

Sjostrand, F. S. 1959. Fine structure of cytoplasm: the organization of membraneous layers. Rev. Mod. Phys. *31*, No. 2, 301–319.

Sonneborn, T. M. 1950. Partner of the Genes. Monogram 39. Scientific American, 11 pp.

Thompson, D'Arcy. 1942. On Growth and Form. Cambridge University Press, Cambridge, England.

Urey, H. C. 1952. The origin of the earth. Sci. Monthly. *187*, No. 4, 53–60.

Waddington, C. H. 1953. How Cells Differentiate. Monogram 45. Scientific American, 7 pp.

Watson, J. D., and Crick, F. H. C. 1953. Genetic implications of the structure of deoxyribonucleic acid. Nature. *171*, 964–967.

Weiss, P. 1959. Cellular dynamics. Rev. Mod. Phys. *31*, No. 1, 11–21.

CHAPTER 12

Radiation Effects on Single Cells

The study of single-celled organisms offers possibilities for their better control and utilization by mankind, and, because a single cell demonstrates the life process, its study also gives insight into the phenomena of the interaction of radiation with all living entities. In the previous chapter, the structure, function, and dynamic processes of cells were discussed. In this chapter, their reaction to being in an ionizing radiation field will be presented, together with the technological importance of the effects produced.

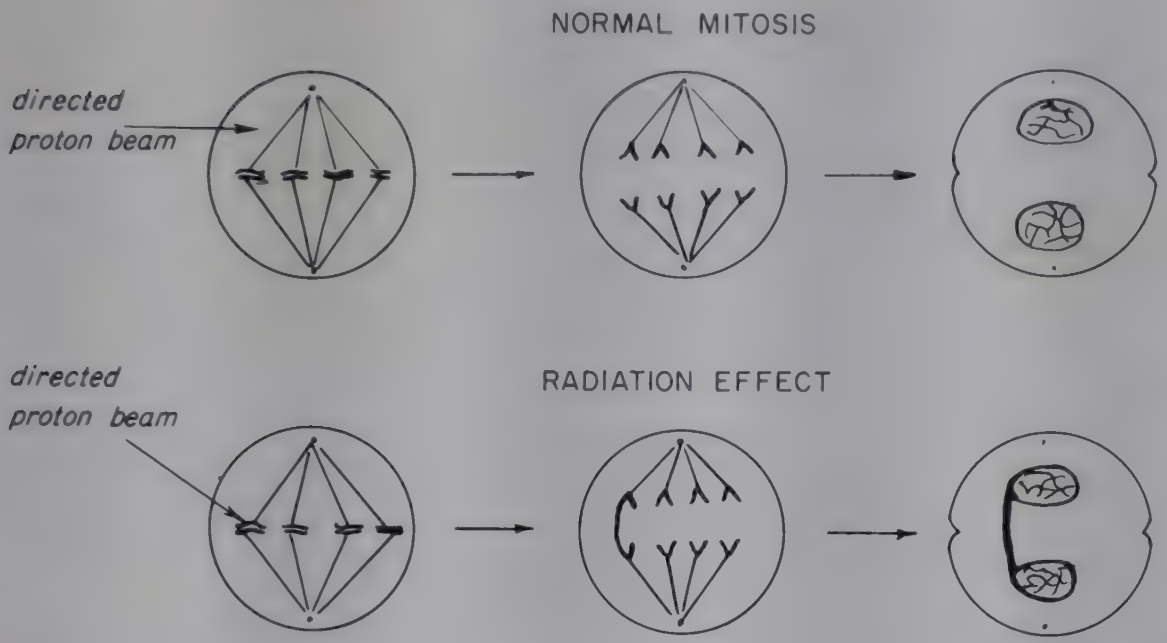
The irradiation of single cells leads to a variety of detectable effects, e.g., gene mutations, changes in membrane permeability, altered nutrient requirements, changes in shape, failure to reproduce successfully, and death. In Chapters 8, 9, and 10, the effect of irradiation on the components of cells was presented. The complications imposed by having these individual biological compounds and polymers organized, oriented, and directed in a successful pattern of life by the organism lead to the above manifestations of radiation damage in the living cell. Those cells which have received the greatest attention have been the bacteria, yeasts, molds, and viruses.

Gene Mutations

While the existence of a nucleus remains to be demonstrated in bacteria, the presence of nuclear material has been confirmed. Viruses are seen as all or mostly nuclear material. Nevertheless, those cells which go through mitosis are found to have definite and rather dramatic responses to radiation exposures. In Figure 58, the mutations which commonly occur in cells were reviewed. It is to be anticipated that as a class these must be capable of occurring in the presence of radiation because life on earth operates in a variable field of ionizing radiations, as discussed in Chapter 1.

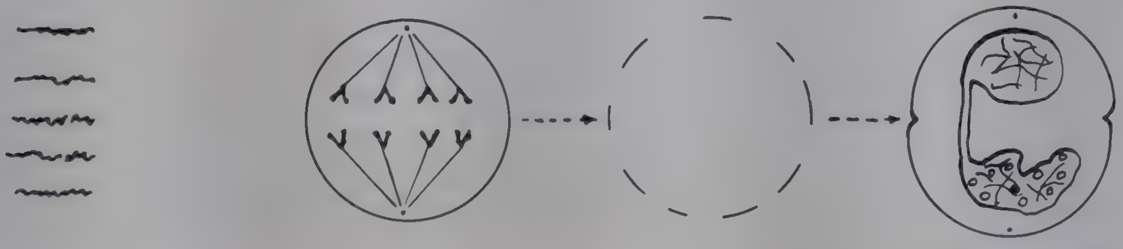
That radiations can indeed cause mutations was shown by the classic work of Muller in 1927. That some of the mutations found in nature are indeed radiation manifestations is shown from the analysis of rates of mutations (**Fig. 59**).

The unexpected feature of the genetical effects due to radiation is that the effect is cumulative—directly proportional to dose, irrespective of time. The irradiation of DNA alters its organic bases (e.g., guanine etc.), and also a DNA particle may be broken into one or more parts. These changes are adequate to give rise to the observed effects of radiation on chromosomes and consequently the genetical effect which they produce and control. Not only are chromosome breaks observed but there is a stickiness which is observed in cell division which does not permit the chromosomes to separate normally, leaving bridges of chromosomal material between them (Bloom 1959). This is shown schematically in Figs. 62 and 63. The spectacular feature shown in Fig. 62 is that a single burst of protons when directed to the area of the spindle of a cell undergoing mitosis does not interrupt the normal execution of the mitotic process although the cell may be injured. In Fig. 63 is shown another situation in which the beam of ionizing radiation has been directed to a chromosome and, from the work of Bloom (1959), it is shown quite clearly to result in incomplete cell nuclear divisions, chromosome bridges re-



Courtesy of Bloom

Fig. 62. Chromosomes Sensitive to Radiation Effects



Courtesy of Bloom

Fig. 63. Radiation Effect on Cell When Exposed to 350 Rads at Anaphase

maining in the frustrated cell. The nuclear component may be bubbled in appearance, not a normal phenomenon.

Beadle and his co-workers (1941), using the fungus *Neurospora*, found that spores given an ionizing radiation treatment are generally killed by the exposure, but those which survive may demonstrate mutations. Usually the survivors have lost the ability to carry out one or more of the chemical reactions in metabolism performed by the untreated spore. It would appear that what has occurred is that in each case a gene has been damaged by the radiation and each gene has been shown to control at least the formation of one enzyme. It is occasionally found that one or more nutrients must be added to the ordinarily suitable basal medium in order to permit the organism to evidence growth. With cross-breeding experiments it has been demonstrated that gene damage is distinct and it is only a matter of chance that two may become damaged in the same spore. Such a damaged gene can be replaced by cross-breeding with a strain which possesses the enzyme which is apparently lacking. It is quite evident that whenever damage has been inflicted on DNA it will clearly interfere with the abilities of the organism. It has been found that ionizing radiation can interrupt the whole sequence of metabolic reactions. If a substance which the organism ordinarily synthesizes cannot now be made due to the absence of the required enzyme, the precursors accumulate since the organism is unable to perform the next step of an essential series. This will be shown later to be a useful phenomenon of value in radiation technology. If the missing ingredient or component is added artificially, the organism may proceed in growth. It is to be anticipated that such exposures lead to alterations in the nutrient demands of organisms as indicated above.

Target Theory and Indirect Action

The biological effects of radiation occur as a result of discrete changes in the atomic and molecular structures of the irradiated material, perhaps less than 0.01 per cent of the chemical bonds being affected, even at several megarad exposures.

Direct Effect.—Biologists have proposed the target theory of direct impact of an ionizing particle with the substrate as being that principally responsible for irradiation effects (Lea 1955). This was based on investigations of biological change, e.g., mutation and lethal effects. The theory proposed that if a swiftly moving charged particle struck a complex of biological material, the biological function of the complex was altered or destroyed.

Within the past decade, evidence has been presented to indicate that the direct "hits" may be responsible for some specific biological effects, but that many effects are caused in whole or part by radiation induced ionization and excitation of the solvent system of the biological material (Zirkle 1959).

Indirect Effect.—As shown in Chapter 8, the irradiation of a water-containing material causes the ionization and excitation of a portion of the water molecules, with the formation of highly reactive hydrogen and hydroxyl radicals. These radicals contribute substantially to the biological effects of ionizing irradiation. Hence, there is an indirect effect of irradiation of moist tissues, caused by these free radicals, due to "activated water."

Hydrogen and hydroxyl radicals are chemically very reactive and can act as reducing and oxidizing agents and also can cleave carbon-to-carbon bonds. Secondary products of irradiation may be of equal importance, since in the presence of dissolved oxygen the hydrogen atom can combine with molecular oxygen to form the very reactive $\cdot O_2H$ peroxide radical,

$$\cdot H + O_2 \rightarrow \cdot O_2H$$

which can form hydrogen peroxide,

$$2\cdot O_2H \rightarrow H_2O_2 + O_2.$$

The hydroxyl radicals may also form hydrogen peroxide,

$$\cdot OH + \cdot OH \rightarrow H_2O_2,$$

The theories of direct and indirect action describe the mechanisms which apply simultaneously in the same system. The indirect theory, however, offers a wider basis for cellular change.

Radiation Effects in Cells

The ability of radiation to kill micro-organisms has been the object of investigation since the late 19th century. While at first negative results were obtained, essentially due to the insensitive techniques used to study the organisms and the low intensities of X-rays employed, it later became clear that ionizing radiations had lethal effects. In 1898 Rieder (Lea 1955) exposed cultures of organisms on agar plates to X-rays and observed definite inactivations. The literature since has been voluminous and the interest in the effects of irradiation on bacteria, yeasts, molds, and viruses has mounted, yet knowledge of the effects of ionizing radiation in living systems is still fragmentary. If we were to follow microscopically a bacterium as it glided through an intense radiation field, we would not see the organism killed as a bullet destroys an animal. Few cells are inactivated outright by the exposure and most retain a portion of certain functions such as motility, respiration, and some biosynthetic activity. When cell division commences, the effects of radiation begin to appear. With a sufficient dose, life processes terminate.

Upon exposure to ionizing radiations, the events which occur in an irradiated cell may be divided into three periods. The beginning period includes the instant of irradiation; the middle period is that time prior to effects being measurable; and finally, the end period is that in which alteration or lesion of the cell manifests itself (Lea 1955).

The beginning period is concerned with the cellular site of the reactions which occur and was considered by early workers in the field to be the sum total of effects of ionizing irradiation. Their reasoning revolved around the theory of gene mutation due to the direct hit or target theory effects. A review of published papers up to 1940 indicates that the principal interpretation of all work was that a definite quantum of radiation must be absorbed within cells for lethality or gene mutation to occur. This conception was modified with the accumulation of data by more refined techniques. The lethal effect can also be due to the passage of ionizing particles either through a cell wall or very near to it. A direct hit in an area less vital than the nucleus may cause injury rather than death.

The middle period of events following irradiation of cells can be thought of as the period during which reactions occur leading to the end effect. During this stage, the cell is in an unusually excited physiological state, which may be indicative of an alteration of its nucleus or perhaps due to the indirect action of radiations upon the cell membranes, the cytoplasm, or even due to activated media surrounding the cell.

The effects of radiation which occur in the end period are, to name a few: lethality, gene mutation, various types of growth inhibition, and alterations in nutrient requirements.

The environmental conditions of living micro-organisms before and after irradiation modify the lethal effects. For example, a greater resistance is found for anaerobic cultures of yeast than for the same organism cultured with vigorous shaking or aeration, the effect being related to the oxygen in the irradiated suspension. The removal of oxygen from suspensions by various methods before irradiation affords protection to the organisms. The presence of catalase, with its specific activity to decompose hydrogen peroxide is also protective and significant because hydrogen peroxide is a product of the irradiation of aqueous systems.

The effects of pre-irradiation and post-irradiation treatments indicate that strains of organisms are affected differently by ionizing radiations (Stapleton *et al.* 1954). This, and other phenomena that have been found, dictates that caution should be exercised in making generalizations concerning lethal as well as genetical effects of ionizing radiations upon organisms. The present state of knowledge is as follows: (a) The direct hit or target theory, which for many years dominated the thinking as regards the effects of ionizing radiation on organisms, does not fully describe the lethal effects of irradiation; (b) Radiation effects have proved to be reversible to a certain extent; (c) The indirect effects of ionizing radiations (the creation of highly excited chemical species) are apparently responsible for killing organisms in many cases.

The Problem of Death

To consider the biological effects of radiation in single-celled organisms, it becomes necessary to establish some basic definitions. Life and death in micro-organisms should be definable in terms other than themselves. Yet, the determination of actual death in a microscopic organism is a difficult problem which leaves the human analyst with only crude methods of expression. Cell division represents a major life process and is a sensitive tool for differentiating between living and dead; yet even this is complicated by the recurrent feature of dormancy.

Death cannot be defined by positive criteria for simple organisms except by the absence of a property called life. Among the essential criteria outlined by Rahn (1945) for death are the loss of motility, or respiration, or of other enzyme activities or the change in permeability of membranes (which cause increased absorption of dyes) all having been used. These features may not appear simultaneously. But some workable objective standard of judgment must be used, and the failure to reproduce is one criterion which is workable.

If we consider cell life as a self-contained, highly organized biochemical system, and at its simplest a series of highly integrated enzymatic processes capable of reproducing in a suitable environment, then death could be conceived as the disorganization of the system, manifested in at least two measurable forms, morphological and physiological. Both are discernable since morphological changes are observable and physiological activity can be measured within a cell and is becoming better understood. While cell division is retarded, a cell may lengthen and swell in size and eventually attempt to divide. Mutants may appear at low doses of radiation, and, with a sufficient exposure, death usually follows when either cell division or growth is attempted. Physiological processes proceed at altered rates, and terminate when enzymes which guide the reactions no longer function, and no replenishment occurs (Pollard 1959). Thus radiation can be expected to have marked and measurable influences on these single-celled organisms.

Order of Destruction of Single Cells

The lethal effects of radiation has received much study and it has been shown that for a variety of organisms the number of survivors measured by reproductive ability decreases exponentially with increasing dose of radiation absorbed. Plotting the logarithm of the number of survivors against dose yields essentially a linear relationship, the slope of which is controlled in large part by the organism and its environment prior to, and during, irradiation. Post-irradiation manipulation of the organism is also an important consideration (Johnson 1959).

Factors Influencing Radiation Destruction of Micro-organisms

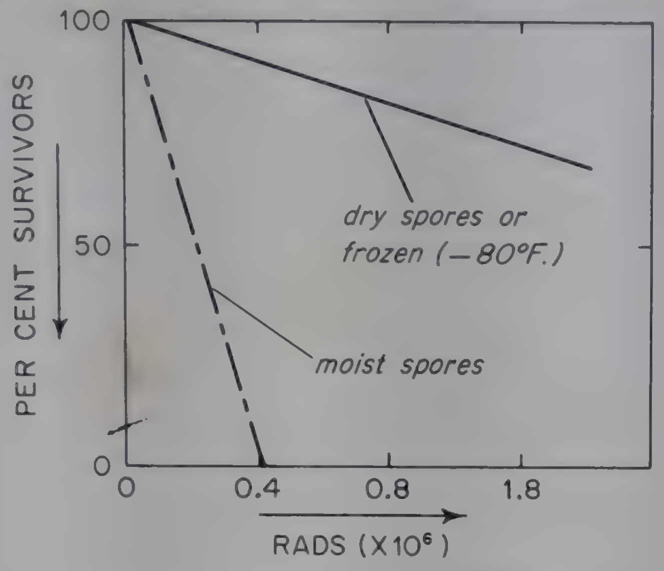
The extent to which an organism is sensitive to radiation is dependent upon the following general factors:

Presence of Oxygen
Presence of Water
Sporulation
Species and different strains of the species
Concentration of organisms
Atmosphere in which irradiated
Physical state of the medium
Hydrogen Ion Concentration (pH)
Composition of medium
Age of culture
Type of Radiation
Postirradiation treatments

Oxygen Effect.—There have been rather marked influences of oxygen in the destruction of organisms with ionizing radiations; these are found to be greater in studies using vegetative forms rather than with spores. In the absence of oxygen, there is a definite protection of vegetative cells, suggesting a radiation induced chemical oxidation, with strong indications of peroxide effects. Catalase possessing organisms are much less sensitive to this oxygen effect. (Williams and Kempe 1957).

Water Effect.—Because of the significance of the radiolysis of water and the production of highly reactive hydrogen and hydroxyl radicals, it is anticipated that the radiation resistance of organisms is greatest in the dry state. Such is, in fact, found to be the case, and this may be seen in Fig. 64. It may be postulated that in the dry state killing of organisms is due mainly to direct hits, while the effects in moist conditions is due to both direct and indirect effects of radiation. A somewhat analogous condition is found in the irradiation of cell suspensions frozen at —80°F., in comparison with exposures at 40°F. In the former case the water molecules exist in the solid state, hence are immobilized, and in the latter case the movement of the free radicals formed is permitted. The ice then acts as a cage for free radicals, which are therefore not free to react except in their immediate locality in the crystal lattice.

Vegetative Cell vs. Spore Radiation Resistance.—As noted previously, an organism capable of forming a spore finds antigenically different proteins in the spore than those present in the vegetative cell. There is also a change in the free-bound water content. It is useful to attempt an explanation of the variation in radiation resistance of these two forms—vegetative and spore—for a species. Increased radiation resistance of the spore state is commonly found in nature over the vegetative growth from which the spore came (Kuhn 1960). For the sake of argument, let us assume the spore to be in a gel and the vegetative cell to be in a sol condition.



Courtesy of Procter

FIG. 64. INFLUENCE OF PRESENCE OF MOISTURE ON RADIATION RESISTANCE OF BACTERIAL SPORES

It has been shown by experiment on the interaction of radiation with matter that the irradiation of a substance produces active species in an extremely nonuniform concentration pattern. For example, a liquid which is irradiated with an arbitrary dose of any of the types of high energy radiation is commonly considered to absorb some particular amount of radiation energy in a unit volume. And it has been found that a significant fraction of the absorbed energy is utilized in the formation of activated molecules, radicals, and ionized species. For purposes of overall considerations of energy and material balance these average values are quite adequate. On the other hand, when one studies the kinetic processes which lead to the formation of end products from the radiation-produced quantity of active species, the microscopic nonuniformity of concentration of these species cannot be neglected. In the case of aqueous solutions, for example, the instantaneous picture of an irradiated solution may be described in the following terms. The liquid is essentially unaffected, except for an occasional spur of highly excited and ionized species. These species in aqueous solutions are considered to consist essentially of hydrogen atoms, hydroxyl radicals, and electronically excited water molecules. If we now consider the chemical reaction of these species with a solute such as an inorganic ion or, loosely speaking, a bacterial cell, it becomes quite apparent that we are dealing with competitive reaction processes in which the microscopic nonuniformity of concentration plays an important role. For example, one of the possible reactions of a hydroxyl radical (discussed in Chapter 8) is to combine with another such radical to form hydrogen peroxide. A second possibility is for the hydroxyl radical to react with the solute ion or a molecule of a cell membrane. Clearly, the relative importance of these two types of reactions depends on two distinct factors. One of these is the probability that, if the two possible reactants are in close proximity to one another, they can react to form a product. The second and qualitatively different consideration is the problem concerning the probability that two reactants can in fact come into close proximity, in which case diffusion is involved.

Again the first of these is the problem of microscopic reaction cross sections or rate constants, and the second is the problem of mobility of the species involved. Because of the way in which active species are initially concentrated in radiation-induced tracks, the reaction between such species is somewhat favored. And this can be dramatically accentuated by making it more difficult for the active species to diffuse over large distances.

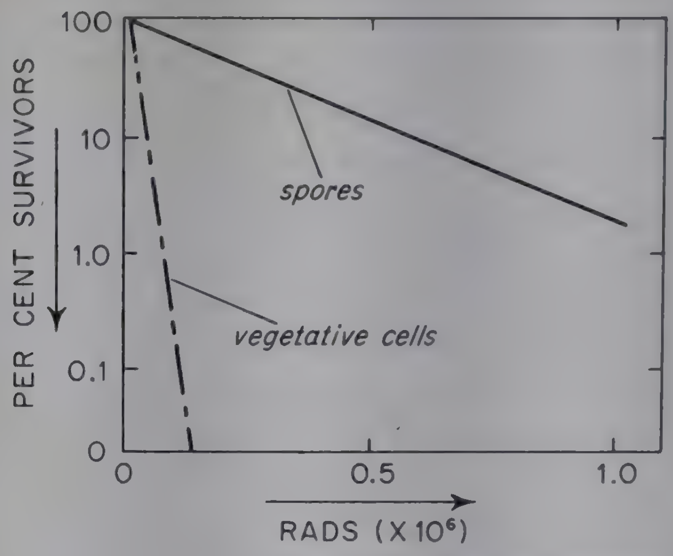
Agar is a substance which can be made a reversible colloidal structure. Heating a suitable water system of agar places it into suspension, and cooling permits the agar to solidify, establishing a rigid gel structure. Heating the gel returns it to a sol condition. It is important to heat the agar above its melting point if a gel is to be permitted to form on cooling. Actually it is possible to have two tubes of the same substrate at almost the same temperature yet have one tube a sol and the other a gel. If bacterial spores are suspended in both solid and liquid agar it is possible to have the spores in chemically identical substrates, which differ in physical state, sol, or gel. The irradiation of such a system might demonstrate whether there is a relationship between rigidity of a substrate, the killing effect of radiation, and the mobility of radicals. To eliminate the temperature effect itself from killing the organisms, we can choose heat resistant bacterial spores which are not injured by exposure to the temperatures studied. The temperature difference between the sol and the gel is a matter of a few degrees and has been shown to have no impact on the lethal effect radiation will have on the organisms. Such an experiment undertaken by the authors does demonstrate in fact that the killing effects are substantially different, the results obtained varying as much between gel and sol condition as dry and wet cells, or cells in ice versus water (Fig. 64).

The difference in agar sol and gel structures can be described in the following terms. The agar molecule is a polymeric carbohydrate of large molecular weight. The hydroxyl group of this polymer molecule can form hydrogen bonds within the molecule and with surrounding molecules. In the sol form the hydrogen bonds tend to be largely intramolecular so that the molecules are somewhat coiled up upon themselves and adjacent molecules may be displaced with respect to one another with little expenditure of force. The preparation of the gel form provides the clue to the structural difference of this system. When the agar sol is heated the increased temperature causes the breaking of some of the intra-molecular hydrogen bonds and allows the molecules to become uncoiled. Upon subsequent cooling, the molecules can now establish intermolecular hydrogen bonds which then impart rigidity to large portions of the liquid. The resulting network structure entraps the water present and its constituents.

If it is considered that the diffusion of any species in water involves the carrying along of immediately neighboring water molecules and exertion of shear forces on the surrounding water, it becomes clear that the entrapment of water on the microscopic scale increases its resistance to such shear forces and hence will impede the diffusion process. It will be recalled that most reactions involve diffusion; however, a decreased diffusion velocity will most seriously affect those processes in which the diffusion length is large. This appears to be the case for reactions involving an active species (free radical) and a suspended particle (bacterial spore) as compared to reactions which can occur among active reactants formed in the same microscopic track where the diffusion length is infinitely small. The fact that diffusion rates differ in agar sol-gel structures is demonstrable with dyes and other agents.

The difference in radiation resistance of bacterial spores and vegetative cells (Fig. 65) requires explanation. Variations in bound water, electrolyte concentration, altered proteins, and many other factors have been suggested to account for the above differences in radiation resistance. However, if it can be assumed that the protoplasm in an actively growing cell is in a sol condition and that of a spore is in a gel condition, it may well be that one of the principal differences in radiation resistance is the mobility of free radicals found inside the cell, perhaps much in the same manner as was demonstrated with the agar sol-gel system. It would appear reasonable that the above is in fact related to destruction by target and indirect effects, target effects predominating in rigid physical structures.

Species.—The most critical factor is the species of organism. There is a wide range of radiation resistance between organisms, and there is a range even between strains within a species. Generally, vegetative cells require substantially less dose applied than spores to accomplish a given degree of kill. The exposure of a concentration of vegetative cells of *Cl. botulinum* to 46,000 rads finds no survivors while the exposure of the same concentration of spores to 400,000 rads may find approximately 10 per cent survivors. Variations within the species have been demonstrated in that Type A spores are nearly twice as resistant as Type B, other factors being equal. The radiation resistance of *Cl. botulinum* spores ranges from 100,000 rads to 400,000 rads to accomplish a reduction in survivors of 90 per cent from initial concentrations (Morgan and Reed 1954).



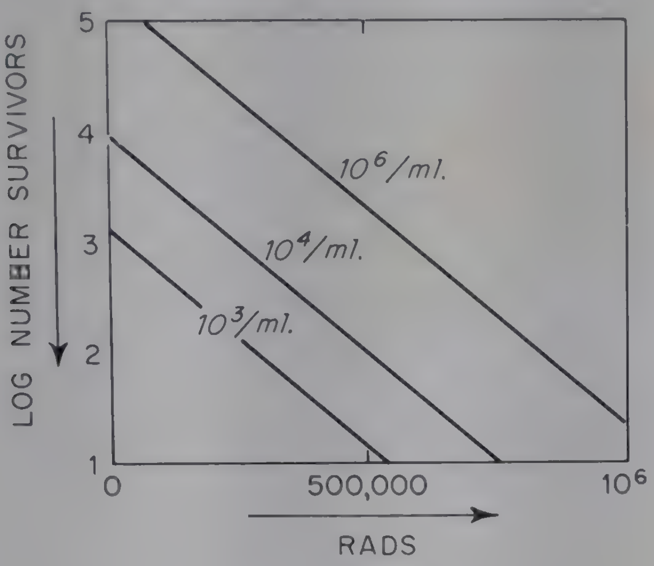
Courtesy of McArdle and Desrosier

FIG. 65. RELATIVE RADIATION RESISTANCE OF SPORES *vs.* VEGETATIVE CELLS OF *Bacillus coagulans*

Cocci and non-spore forming rods are generally radiation sensitive. Population reductions of 90 per cent with *E. coli* may be obtained with as little as 20,000 rads.

Concentration.—Death to irradiated cells appears exponentially and the proportion of survivors with increasing increments of radiation treatment are generally predictable. As seen from Figure 66, while the proportion of survivors is a regular feature, the actual number of survivors is dependent upon the initial concentration. A 90 per cent reduction (one *D* value) of a population containing 1,000,000 spores finds 100,000 survivors. A 90 per cent reduction in a population of 100 spores leaves 10 surviving. To bring a substrate to a final concentration of organisms, it is necessary to know the initial population density.

Atmosphere.—As indicated earlier, the irradiation of water leads to the production of reactive hydrogen and hydroxyl radicals. In the presence of oxygen, the yield of hydrogen peroxide is increased, and its killing effects are well recognized. The exposure of vegetative organisms in the absence of air finds less destruction to living cells than the exposure under equal conditions except for the presence of oxygen, the oxygen effect being detectable in the latter case. This effect is not found for nitrogen or other gases. Bacterial spores do not show as marked effects as do vegetative forms as noted earlier.



Courtesy of Billerbeck and Desrosier

FIG. 66. INFLUENCE OF CONCENTRATION IN RADIATION RESISTANCE OF *Bacillus subtilis* SPORES IN NEUTRAL BUFFER SOLUTION

Physical State of Medium.—As previously discussed, the fluidity of the irradiating substrate may have significant effects on the ability of the treatment to destroy cells. When free radical diffusion is retarded or prohibited, the indirect effects of ionizing radiation will not function, hence larger doses are required for an equal kill.

In addition to the physical state of the substrate, it is important to consider the temperature at which irradiation occurs. Earlier work was undertaken with frozen samples, the destruction of cells being mainly by direct hits. On the other hand, elevated temperatures offer promise in increasing the kill of organisms.

It is well recognized that with temperatures in the vicinity of that of boiling water, organisms, especially vegetative forms, are readily killed without radiation. Interestingly, it has been found that the exposure of bacterial spores to low levels of ionizing radiation sensitizes the cells to destruction by further low heating, reductions in heat resistance being in the order of five fold or more.

It would appear that there is a temperature condition in which the irradiation of cells would yield maximum death to a population and this has been found in several instances. There would appear an optimum temperature for irradiating cells in the range of 120–160°F.

Hydrogen Ion Concentration.—While the organisms which are detrimental to man grow in a rather narrow range of pH values, there is little information pertaining to the effect of pH alone in influencing the killing effect of ionizing radiations. If the effect occurs, it is of low order of importance, especially through the useful range of 4.0 to 7.5.

Composition of Medium.—The substrate in which cells are exposed has marked influence on the destruction of cells. Protein in the medium is protective to cells, and consequently higher dose levels are required to destroy cells in the presence of protein than in a neutral buffer solution or in distilled water, see Table 28.

TABLE 28

SURVIVAL OF STAPHYLOCOCCUS AUREUS IN WATER, DILUTE GELATIN, AND NUTRIENT BROTH[1]

Substrate	Per cent Cells Surviving Irradiation Dose of 20,000 Rads
Water	12
Dilute gelatin	22
Nutrient broth	42

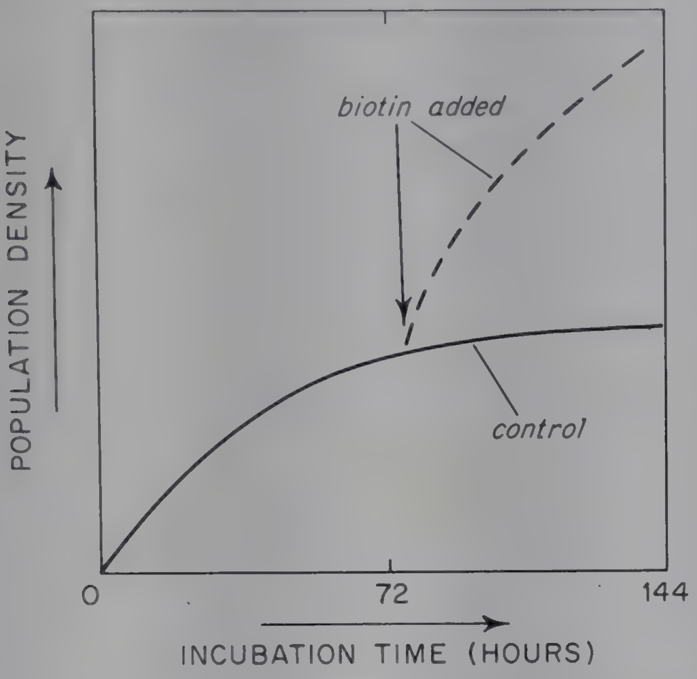
[1] From Hollaender and Stapleton (1954)

On the practical side, the *D* values for *Cl. botulinum* in cream of chicken soup, whole kernel corn, and boiled ground beef were reported found to be 0.352, 0.358, and 0.348 million rads respectively, based on incubation tests with inoculated cans, using a million spores per can. The 12 *D* value level would be 4.2, 4.3, and 4.2 million rads, respectively. Pea purée on the other hand has a *D* value of 0.330 and a 12 *D* value of 4.0 million rads (Anellis and Huber 1958). There is a slight variation between media, but of relatively minor consequence. The presence of water is highly significant, as shown in Fig. 64.

A large number of chemical additives are protective to cells, including sodium hydrosulfite, sulfhydryl compounds, alcohols, aldehydes, glycols, carboxylic acids, and sugars. Cysteine has pronounced protective effects.

It is to be expected that compounds capable of reacting with free radicals will be protective to cells. The effect of reducing and non-reducing sugars in various concentrations and their protective effect to bacterial spores has received attention, glucose actively reacting with ·OH. While more than ten times the survivors appear in sugar solutions, more than 90 per cent of the spores are killed by 500,000 rads. A one molar solution is nearly 20 per cent sugar.

Age of Culture.—Young cells appear more radiation resistant than aged cells, although the effect is not clear with spores. Considering this factor is generally not one easily controlled, it is therefore a minor consideration.



Courtesy of Johnson and Desrosier

FIG. 67. EFFECT OF ADDED BIOTIN ON GROWTH OF IRRADIATED SPORES *(Bacillus coagulans)*

Maximum resistance has been found at the end of the lag phase of growth, an increased sensitivity to irradiation during the logarithmic phase, followed by a gradual recovery of resistance in the stationary phase of growth.

Type of Radiation.—The work of Proctor and Goldblith (1952) on the germicidal efficiencies of gamma rays, X-rays, and cathode rays has indicated that the effects of these radiations are essentially identical when evaluated in terms of the killing effect of equal doses.

The fact that bacterial spores can be killed by heat and by irradiation has led to speculation on the similarities and differences in alteration of metabolic pathways in each instance. Although no clearly defined direct enzymatic relationship has been demonstrated, it appears that there is

TABLE 29

VIABILITY OF GAMMA-IRRADIATED B. COAGULANS NRS NO. 770 SPORES HELD AT 3 °C.

	Weeks				
Rads	0	1	2	4	6
0	1,300,000	900,000	700,000	800,000	900,000
7,500	180,000	120,000	120,000	130,000	140,000
20,000	100,000	50,000	63,000	67,000	72,000
35,000	52,000	19,000	30,000	25,000	36,000
53,000	20,000	12,000	12,000	15,000	15,000
100,000	2,700	1,300	930	1,200	1,100

an involvement responsible for the difference noted between the heat and gamma irradiation treatments. This is based on the known effects of both agents on enzymes. Heat has certainly been shown to be far more destructive of an enzyme complex than irradiation. In studies in which a given per cent kill is accomplished with a set of spore suspensions of the same culture by heat and by irradiation, metabolic studies do indicate slight differences in the ability of survivors to utilize single sources of energy, but the differences do not explain the anticipated variations. In part it might be argued that heat gives a more constant treatment than irradiation, but that the nature of the destruction curves by each method are quite similar. Perhaps the explanation is to be found in the impaired ability of cells to synthesize enzymes.

For another problem, evidence the nutrient demand of irradiated cells over that of control cultures shown clearly in Fig. 67. The addition of biotin to irradiated spores of *B. coagulans* manifests itself in rather spectacular growth when compared with nonirradiated controls.

Recovery of residual spores following irradiation over a six week period (see Table 29) has shown that the number of recoverable spores after a one week holding period falls off to approximately one half the number recoverable shortly after irradiation. On continued holding, the number of viable spores increases again (Billerbeck 1959).

TABLE 30

GAMMA RADIATION-INDUCED HEAT SENSITIVITY IN SPORES OF B. COAGULANS NRS NO. 770 AND THE INFLUENCE OF POST IRRADIATION STORAGE AT 3 °C.[1]

	Weeks				
Rads	0	1	2	4	6
0	8[1]	5	3	1	0
7,500	6	22	17	15	17
20,000	8	24	13	27	17
35,000	23	50	17	24	35
53,000	25	33	36	22	32
100,000	26	26	31	37	33

[1] Per cent reduction in numbers following heating 10 minutes at 80°C.

Spores of *B. coagulans* are more heat sensitive following irradiation at sub-lethal levels (Table 30). The magnitude of this induced sensitivity is appreciable. Non-irradiated spores following heating for ten minutes at 160° F. yields a negligible reduction in populations over those unheated. Irradiated spores similarly evaluated indicate that the remaining spores are at least several orders of magnitude more sensitive than unirradiated controls. No change in the induced sensitivity is found upon holding the treated spores for six weeks. Heating followed by irradiation does not yield this result, based on present data.

Investigation into the nature of death to cells metabolically is warranted. "Inactivation" might be a more appropriate term than"death" which carries with it a note of finality and irreversibility.

Reversal of Damaging Effects of Ionizing Irradiations

A partial reversal of damaging effects of ionizing radiations has been demonstrated by Stapleton *et al.*, (1954) with *Escherichia coli* (a fecal organism) cultured aerobically in nutrient broth and plated after being subjected to X and gamma irradiation on the same medium. Maximal survival was observed when the organisms were incubated at a temperature well below that which is optimal for growth of this organism. The radiation apparently induced an altered optimal temperature for colony formation. This suggests that the temperature at which irradiated cells will best survive may be determined by the relative rates of synthetic and destructive processes in these cells, and since the irradiation apparently induced an alteration in the optimal temperature for colony formation, it seemed reasonable to expect that the irradiated cells might also become more fastidious in their nutritional requirements.

For example, *E. coli* grows equally well on a simple synthetic medium composed of inorganic salts with a carbon source such as glucose or on natural materials such as nutrient broth. The comparative viability of irradiated bacteria plated on synthetic media of several types and on a complete medium such as nutrient agar or yeast extract, demonstrates that a lesser number of the cells apparently survive when cultured in nutrient broth as compared to those plated on a synthetic medium. The addition of known useful organic compounds to a synthetic medium were conducive to recovery. Holding an irradiated suspension of *E. coli* in the presence of yeast extract at 98.6°F. for 30 minutes results in the later formation of equal numbers of colonies on non-supplemented and supplemented media. Evidently some factor(s) in yeast actually stimulate repair mechanisms in the irradiated cells. The addition of biotin and other vitamins to recovery media further demonstrates this concept (Johnson 1959).

Post-irradiation treatments with metabolic inhibitors (arsenate, azide, and dinitrophenol), and various cell nutrients (N-sources and sugars) indicate that the biological effects of irradiation can be modified by the post-irradiation metabolism.

There appear two requirements for the recovery of *E. coli* cells from irradiation: (a) incubation at sub-optimum temperatures, and (b) the presence of a certain nutrient substance. Hollaender (1954) found that a medium composed of inorganic salts and glucose supported growth of non-irradiated cells but was unquestionably limited for the irradiated ones, and found it essential to add either yeast or meat extract. A synthetic mixture of amino acids and known vitamins and growth factors permitted slight recovery but these did not approach the effectiveness of yeast or meat extract in the recovery process. Of the various tissues of the animals, spleen extracts contain the factor in highest concentration.

It would appear that the recovery process of a cell from ionizing irradiations is related to the synthesis of new enzymes in the irradiated cell. Further, it appears that the mechanism for biosynthesis of enzymes has been injured, in relation to the dose received. On the basis of meager information, it might be suggested that direct effects (target) of ionizing radiation on a cell cause death in a different manner than death due to the indirect effects. Direct hits on a critical system in biosynthesis and reproduction have been conceived by Lea (1955) as follows: The typical effect of one ionization is chemical change in one molecule, and it is unlikely that the concentration of any cytoplasmic constituent is so nicely balanced that change in a single molecule of it leads to the death of the bacterium. On the other hand, in organisms which have been investigated genetically, lethal effects due to losses of, or changes in, single genes are common.

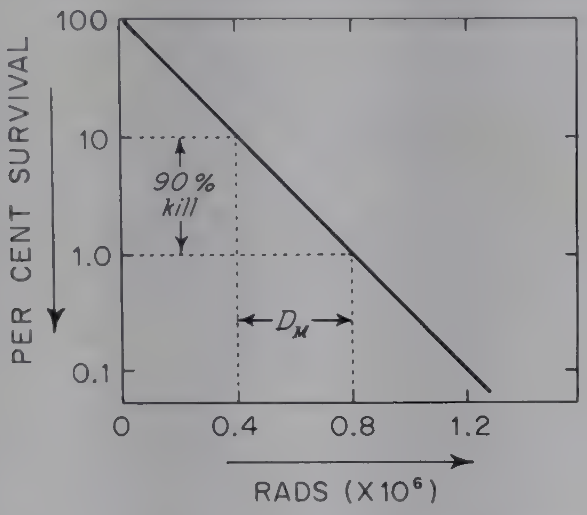
In research on which is based the conclusion that death is due to a single ionization, the criterion of death has invariably been the inability of the irradiated bacterium to give rise to a colony visible to the naked eye or under low magnification. The microscopical studies which have been made of the behavior of individual bacteria after irradiation have shown that a bacterium exposed to a dose sufficient to prevent its giving rise to a colony does not suffer rapid disintegration as happens for example when bacteria are killed by strong chemical disinfectants. A motile bacterium retains its motility (Bruynoghe and Mund 1925), and though failing to give rise to a visible colony, a bacterium which has been "killed" by radiation may divide once or twice when sown onto a suitable nutrient medium (Luria 1939). A spore which has received a dose large enough to prevent its giving rise to a visible colony might nevertheless germinate when sown onto an enriched nutrient medium. These results suggest that, as is often the case with higher cells, the death of an irradiated bacterium occurs at or following division. The cause may very well be an effect of radiation on the genetical apparatus.

On the other hand, indirect effects may be manifested in altered cell wall permeability. Here perhaps the principal effect could be the altered ability of the membranes to assimilate nutrients from a medium, not all essential molecules being permeable after the cell membrane has been distorted by interaction with free radicals.

A complete understanding is still not available.

Radiation Destruction of Bacteria

Bacteria are commonly referred to as the simplest members of the plant kingdom, and exist as one-celled entities. These cells may occur as single spherical units, as rods, or as spores. The rods may be found in chains and the spheres may be found in strings or in clumps, features which are useful for classification purposes. Bacteria are separated on the basis of morphological and physiological characteristics. In addition, some bacteria have the ability of forming spores. The vegetative or growth forms of bacteria are generally not resistant to heat or the action of chemical agents, and therefore it may be anticipated that they will have relatively little resistance to ionizing radiations, which turns out to be the case. From the standpoint of sterilization processes, the radiation resistance of bacterial spores is of special interest (Fig. 68).



From Desrosier

FIG. 68. RADIATION DESTRUCTION OF BACTERIAL SPORES—THE D_M VALUE—FOR *Clostridium botulinum*

There are two important genera of bacteria which form spores; both genera are rod forms, one (*Bacillus*) is aerobic, and the other (*Clostridium*) is anaerobic. When a rod is about to sporulate, a tiny refractile granule appears in the cell. The granule enlarges, becomes glassy and transparent, and now resists the penetration of various chemical substances. All of the protoplasm of the rod seems to condense into the granule, or young spore, in a hard dehydrated, resistant state. The empty cell membrane of the bacterium may separate off, like the hull of a seed, leaving the spore as a free, round, or oval body. Actually a spore is an end product of a directed series of enzymatic processes. There is no unanimity of opinion either of spore function in nature or of the factors concerned in spore formation.

Since no multiplication takes place as a result of the vegetative cell-spore-vegetative cell cycle, few microbiologists accept the concept of the spore as a cell set apart for reproduction. Instead, various explanations of the biological nature and function of bacterial spores have been advanced, including the interpretation of the spore as a resistant structure produced to enable the organism to survive an unfavorable environment, the idea that the spore is a normal resting state (a form of hibernation), the notion that spores are stages in a development cycle of certain organisms, or a provision for the rearrangement of nuclear material. It is interesting to note that the protein of the vegetative cell and the protein of the spore may be antigenically different.

Spores appear to be formed by healthy cells facing starvation. Certain chemical agents (glutamic acid) may inhibit the development of spores. No doubt sporulation consists of a sequence of integrated biochemical reactions, and the sequence can be interrupted at certain susceptible stages.

The literature on the subject of the radiation resistance of bacteria contains many contradictions and discrepancies from the records of the earliest workers to those of the present day. This lack of uniformity has been due in part to factors of unknown nature. Until the factors operative in the resistance of cells are understood, it will not be possible to control by other than empirical means the processes which require their destruction for success.

While the relative killing effects of the various ionizing radiations available have been the subject of much discussion, it remains that there are two major sources of ionizing radiation of importance and available which can be applied practically in sterilization, these two being high energy electrons (or beta rays) and gamma rays. Evidence is available which shows that the killing effects of both are very similar and that for all practical purposes it may be assumed that a sterilizing dose for one form is equal to that of the other, both being useful for this purpose (Goldblith and Proctor 1954).

The destruction of bacterial spores with ionizing radiation finds the number of cells killed by absorption of equal successive units of dose to be unequal; the relative proportion of survivors after absorbing a unit dose, however, is the same. This is best shown in diagram, as in Fig. 68, for *Clostridium botulinum* in a beef flesh extract. The number of survivors is seen as an exponential expression, and explained in large part by Lea, (1955). While this would indicate that a definite action is required to destroy the organism rather than an accumulated effect, it could still be argued that if the cells in a suspension were of uneven resistance, and killed by a cumulative action, the same type curve could be obtained. The survivors of radiation exposures of high doses, several million rads, when subcultured, do not yield suspensions of notably increased radiation resistance. The survivors are perhaps best considered in terms of the probability of an organism surviving and its increased chances of being sufficiently disorganized as the dose increases.

If the effects were cumulative, the survival curves would be sigmoidal. If the action were a single action type, an exponential shape would be expected, and this is usually found.

Occasionally there is a "tailing off" of the destruction curve, and this in part may be explained by the non-homogeneity of resistance in a culture, which may be due to a number of factors. Whatever the mechanism involved in inducing curvilinear destruction curves, similar problems are found in heat destruction of organisms, and it would appear that the same class of factors may be concerned (Billerbeck 1959).

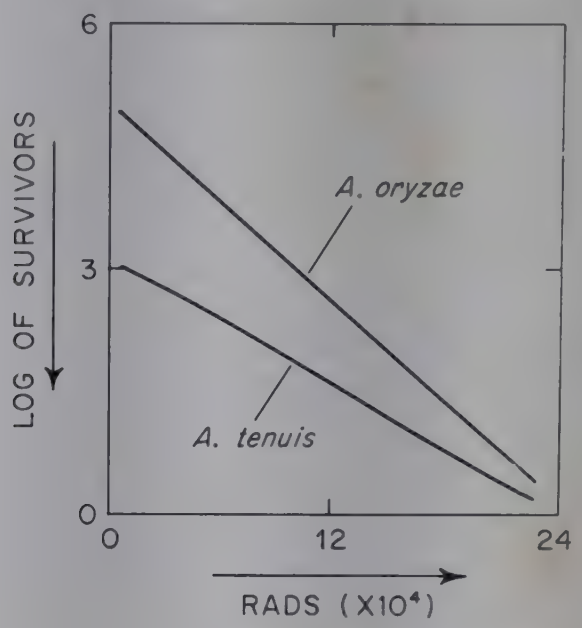
When organisms exhibit a tendency to clump and counting is done by plating techniques, each clump will give but one colony, irrespective of the numbers of individual cells present. This would lead to apparent sigmoidal survival curves, yet this phenomenon alone does not explain satisfactorily the curvilinear responses often obtained.

These complications make it difficult at times to obtain numerical values for rates of bacterial destruction, not only in radiation studies, but in others as well.

Destruction of Molds

The irradiation of molds as a group has received relatively little attention in comparison with bacteria. Molds have less public health significance, hence this is perhaps one reason for the lag in investigation. Yet, molds inflict damage and destruction to marketable commodities estimated as nearly seven per cent of the production of all farm and forest growths. In addition to plant pathogens, there are numerous fungi which act as secondary invaders. The *Aspergillus flavus–oryzae* group, of the latter type, is world-wide in distribution and omnivorous in substrates upon which they are able to grow. This organism is then a useful test species.

A. oryzae is killed exponentially by ionizing radiations; sometimes a break in the curve is found indicating variability in the resistance within the population. Both the plate count method and the spore germination method show that *A. tamarii* is more resistant than *A. flavus,* with *A. niger* intermediate in resistance. The death curve of a *Fusarium sp.* has been found to be sigmoidal, the organism having multi-cellular spores (Fields 1959).



Courtesy of Burns and Desrosier

FIG. 69. RADIATION DESTRUCTION OF FRUIT PATHOGENS

The rate of destruction for typical mold spores is shown in Fig. 69. Exposures at a million rad level ordinarily yield no survivors, yet occasionally even at high dosages a cell might begin to divide or make a division or two before swelling and dying (Burns 1956).

The molds are aerobes. Irradiation in the presence of oxygen increases the kill, while treatments in its absence have reduced killing effects.

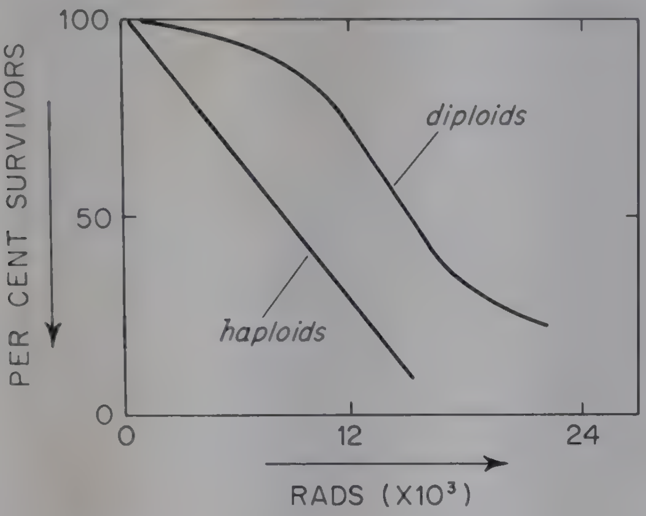
Inactivation of amylase in the cells appears to be associated with growth inhibition of the fungus, while sucrase activity is less sensitive to radiation damage.

The growth rate of *A. oryzae,* measured in terms of dry mycelial weight, is somewhat greater when irradiated starch is the sole source of carbon, especially when the starch had been exposed to extremely high dosages, in which case the data indicate a tendency toward a higher growth rate.

Destruction of Yeast

The exposure of yeasts to 10,000 rads does not find them all killed outright, but daughter cells appear, are unable to multiply, then die. Greater doses kill the cells outright.

The killing curves depend on the nuclear condition of the yeast. Haploid yeasts have one set of chromosomes, diploids two, and polyploids multiple sets. The destruction of such yeasts requires either one, two, or multiple target areas to be destroyed, and such a requirement is shown in their rates of destruction for a given dose. The haploids exhibit exponential killing curves, the diploids take a sigmoidal function and multiploids demonstrate more curvature; the first two are shown in Fig. 70.



Courtesy of and adapted from Tischer and Kurtz

FIG. 70. DESTRUCTION OF MOIST YEAST CELLS WITH IRRADIATION

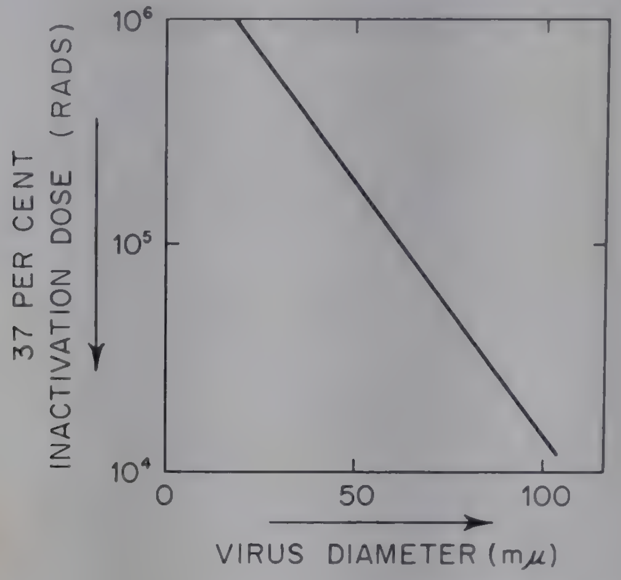
At room temperature approximately twice the radiation dose is required to kill a suspension of *Saccharomyces cereviseae* in the absence of oxygen as in its presence (Lea 1955).

The irradiation of dry yeasts is less destructive for a given dose than irradiation in moist conditions.

Destruction of Viruses

Viruses are distinguished from bacteria and other micro-organisms by their small size and purely parasitic characteristics. Included within the broad category of viruses are the bacteriophages, specific in living in or on bacterial cells. Viruses and phages at present have not been made to multiply on non-living substrates, although tissue cultures are acceptable to them. These organisms in some cases have been crystallized, and the crystalline viruses are chemically mostly nucleoproteins. The smallest crystalline viruses are chemically not dissimilar from genes, and it is possible perhaps to consider a virus as a naked gene structure. Large viruses have not been crystallized and perhaps are, in fact, minute cells; with electron microscopy it has been shown that other constituents are present in addition to the nucleoprotein structure.

Lea (1955) found that the inactivation of a virus with radiation was an action to which the target theory was applicable, and that target size should be related to an experimental inactivation dose. Such relationships are shown in Fig. 71, where destruction of 37 per cent of the viruses was the criterion.



Courtesy of Lea

FIG. 71. RELATION BETWEEN VIRUS DIAMETER AND DOSE REQUIRED TO INACTIVATE 37 PER CENT OF VIRUS

The relationship between virus size and radiation resistance could be considered to mean that a virus contains radiosensitive and radioinsensitive components. Radioinsensitive components would mean those in which ionization produced chemical changes without loss of viral infectivity, which component is radiosensitive. In a virus, the radiosensitive material need not be concentrated in a single mass or area but could be a lamina or a filament. Electron micrographs of the larger viruses show internal structure, indicating that the composition throughout the cell is not uniform, and could well be differentiated into sensitive (or infective) and insensitive components. In crystallizable viruses no such differentiation in radiosensitivity appears to exist. In large viruses the decreasing radiation resistance has led researchers to conclude that there is a differentiation between radiosensitive and insensitive material, more closely resembling higher cells.

Viral inactivation can be caused by direct or indirect effects of radiation, the latter reaching the cell from a track which does not penetrate the cell. At least two types of indirect effects can be identified, one which destroys the ability to adsorb onto a host bacterium and another effect which persists after the irradiation treatment, probably due to extended life of free radicals. Inorganic salts in water solutions are protective to viruses, sodium, and ammonium nitrates reportedly giving some 5,000-fold protection, for example. The pH of the irradiation medium influences the destruction of viruses, as does oxygen's presence.

Protein molecules in the irradiation medium are protective to viral destruction, as are cysteine and glutathione. Their presence offers nearly complete protection against what otherwise would be an inactivation dose. The presence of the enzyme catalase has a two-fold protective action, one in being a large protein structure and second in its specific action of decomposing hydrogen peroxide. The radiation resistance (*D* value) of a number of viruses in several substrates is shown in Table 31.

TABLE 31

RADIATION KILLING DOSES FOR SELECTED VIRUSES

Species	D^1 value (Rads) ($\times 10^6$)	Substrate
Vaccinia	0.4	10 per cent testicle emulsion
Rabies	0.5	10 per cent brain emulsion
Influenza, human	0.4	alcohol precipitate
Newcastle	0.7	allantoic fluid
Polio—Lansing	1.4	25 per cent cord emulsion
Staph V phage	0.3	broth
T-2 phage	0.4	broth
T-3 phage	0.6	broth

[1] *D* value—dose required to reduce population by 90 per cent.

It is interesting to note that from 40–100 ionizations have been calculated to be required before one inactivating event occurs in a virus (Lea 1955). With less ionizing events, injury rather than death occurs. Aside from the application areas in killing viruses, it would appear that ionizing radiations are unique as a tool used to study certain fundamental host-virus relationships and can contribute to the understanding of viral morphology as well.

Technological Importance of Radiation Effects

Sterilization.—Ionizing radiations are unique in their ability to produce chemically reactive species in cells without heating the substrate to any significant degree. This has important technological ramifications. It has been shown earlier that the destruction of cells with radiation occurs in a predictable fashion, and the killing effect may be manipulated by altering the oxygen tension, the amount of water present, and so forth.

The lethal effect of ionizing radiation on a suspension is usefully expressed in terms of the decimal reduction dose required to reduce the original population by 90 per cent. The D_M value, as it is then called, is independent of concentration, see Fig. 66; the number of survivors in a population is concentration dependent, as seen in the figure. The function has been expressed as follows (Rayman and Byrne 1957):

$$D = D_M (\log N_0 - \log N)$$

where D = desired level of destruction
D_M = is the 90 per cent reduction value
N_0 = is the initial population
N = final concentration required.

If the destruction curve shown in Fig. 68, had been plotted as the logarithm of the number of survivors, when the curve has traversed one log cycle the bacterial population would be mathematically decimated. The D_M value can be taken directly from such a curve.

An example of its use may be seen as follows. Assume a D_M value for an organism to be 400,000 rads, the initial concentration of organisms to be 1,000,000 per unit container, and that in 10^6 units no survivors should be found in any of the units, or less than one organism should be found per 10^6 units. N then equals 10^{-6}. By substituting in the equation above,

$$\begin{aligned} D &= 400{,}000\ [6 - (-6)] \\ D &= 400{,}000\ (12) \\ D &= 4{,}800{,}000 \end{aligned}$$

The dose required to achieve this degree of sterility would be 4.8 megarads, for this organism under these circumstances.

The destruction curve does not always pass through the origin. A lag in killing effect is sometimes found, especially at low dose levels, and is dependent on the organism, the substrate used for exposing the organism, and other factors. It may be desired to correct for the lag factor. If the lag is defined as L rads, then a corrected equation above would read:

$$D = D_M (\log N_0 - \log N) + L\ .$$

The application of radiation to the destruction of micro-organisms and other single-celled parasites has a multitude of possibilities. The sterilization and disinfection of foodstuffs alone offers sufficient promise to warrant its exploitation (Desrosier 1958). Then, too, the sterilization of drugs and pharmaceutical preparations is of substantial industrial interest. Drugs, bandages, ointments, plastic tubing, and sutures for surgical uses are examples. A wide range of biological materials have been studied, including blood vessels, skin, bones and bone homogenates, blood, serum, and preparations. The treatment of cadavers should be included. The use of ionizing radiations in sterilizing plastic films for packaging problems where sterility is required is promisng. Many heat sensitive plastic materials are satisfactorily sterilized with radiation.

Application areas exist in controlling the molding of clothing, tenting, canvas, textiles, and leather products by suitable packaging and brief exposure to ionizing radiations. This may be of substantial import in tropical areas where controlling mildew generally is difficult because products are stored in moist, warm conditions.

The destruction of parasites in water supplies, such as amoebic dysentary organisms, which are resistant to chlorine treatments, could make substantial contributions to public health. Sewage disposal problems, including night soil control, are important in controlling the spread of diseases.

Low-level irradiation of meat animals for parasite control is within the grasp of present technology. Inexpensive control is possible and could be applied almost immediately in eliminating certain infections of man by those parasites transmitted through water and food supplies. (See Table 37, pp. 268–269).

The ability of radiation to kill viruses without heating and without subsequent physical damage to the viral entity makes this application of radiation one of substantial interest in disinfection of food and biologicals and also in the preparation of vaccines.

Desirable Alterations in Cell Metabolism through Irradiation.—The metabolism of yeasts cells has received substantial attention, leading to useful irradiation processes. Exposure of yeasts to 80,000 rads has been found to increase their ergosterol content by 200 per cent. Meissel has suggested that the increased yield of ergosterol is an attempt by the cell to adjust to the oxidation-reduction conditions within the cell. The process is most useful in the subsequent formation of vitamin D, and is an example of useful irradiation manipulations of organisms. This area of application has barely received attention, yet the potential appears most promising.

Medical Radiation Therapy.—The application of directed beams of ionizing radiation to the destruction of cells, especially in carcinogenesis, has found widespread use for the benefit of man. The ability to alter the radiation sensitivity of cells with oxygen effects and/or increasing electron density per unit area to be treated by impregnation with metal particles, and/or the addition of protective substances such as cysteine to cells to be protected during irradiation, and/or altering the fluidity of the cell masses and other techniques are seen to lead to more intensive specialized uses of radiations. The dose required to destroy cancerous cells is found to be in the same range (7,500 rads) as that required to interrupt the growth of plant cells, and will be discussed in a later chapter.

Useful Mutations.—The ability of ionizing radiations to induce mutations in cells has a promising future in terms of the technological opportunities in developing useful new strains of organisms. Considering the cell as a remarkable entity with wide capabilities of synthesis, the alterations of organisms to yield specific and useful metabolic products appears most promising.

BIBLIOGRAPHY

ALLEN, A. O. 1954. The yield of free H and OH in the irradiation of water. Radiation Research *1*, 85–96.

ANELLIS, A., and HUBER, D. A. 1958. Determination of the differences in resistance of *Cl. botulinium* types and strains to ionizing radiations. U. S. Army Quartermaster Food and Container Institute, Chicago, Illinois, Research Project Rept. 20–58.

BACQ, Z. M., and ALEXANDER, P. 1955. Fundamentals of Radiobiology. Academic Press, Inc., New York.

BEADLE, G. W., and TATUM, E. L. 1941 genetical control of biochemical reactions of *neurospora*. Proc. Nat. Acad. Sci. *27*, 499–506.

BERAHA, L., and RAMSEY, G. B. 1958. Control of pest harvest diseases of fruits and vegetables by radiation treatment. U. S. Army Quartermaster Food and Container Institute, Chicago, Illinois, Research Project S-525, Report No. 4.

BILLERBECK, F. W. 1959. Effect of equal lethal radiation and thermal treatments on bacterial spore germination. Ph.D. Dissertation, Purdue University, Lafayette, Indiana.

BLOOM, W. 1959. Cellular responses. Rev. Mod. Phys. *31*, No. 1, 21–29.

BRUES, A. M. 1956. Modes of radiation injury: bone and carcinogenesis; and sterility and foetal damage. Progress in Nuclear Energy, Series IV, Biological Sciences. Pergamon Press, New York.

BURNS, E. E. 1956. Maturation changes in tomato fruits induced by ionizing radiations. Ph.D. Dissertation, Purdue University, Lafayette, Indiana.

BUTLER, J. A. V. 1959. Inside the Living Cell. Basic Books, Inc., New York.

DESROSIER, N. W. 1958. Food irradiation research and its practical application in Europe and the United States. The European Productivity Agency, The Organization for European Economic Cooperation, 2 Rue Andre Pascal, Paris 16, France.

DESROSIER, N. W. 1959. The Technology of Food Preservation. Avi Publishing Company, Westport, Connecticut.

DESROSIER, N. W., ROSENSTOCK, H. M., and KUHN, G. 1959. Radiation destruction of organisms in sol-gel systems. Purdue University, Lafayette, Indiana.

FIELDS, M. L. 1959. Effect of cathode rays on food spoilage fungi. Ph.D. Dissertation, Purdue University, Lafayette, Indiana.

GORESLINE, H. E., and DESROSIER, N. W. 1959. Preservation of foods by irradiation. Amer. J. Public Health *49*, No. 14, 488–493.

GOLDBLITH, S. A., and PROCTOR, B. E. 1954. Relative merits of cathode rays and gamma radiations. Nucleonics *12*, No. 2, 32–35.

GOLDBLITH, S. A., PROCTOR, B. E., DAVISON, S., KAN, B., LANG, D. A., BATES, C. S., KAREL, M., and OBERLE, E. M. 1953. Comparative bactericidal effects of three types of high energy radiations. Food Technol. 7, Supplement p. 27.

HOLLAENDER, A. 1954. Radiation Biology. McGraw Hill Book Co., New York.

HOLLAENDER, A., and STAPLETON, G. E. 1954. A symposium on the chemistry of biological aftereffects on ultra-violet and ionizing radiations. British J. of Radiol. *27*, 117–121.

JOHNSON, K. R. 1959. Some nutritional aspects of irradiated and nonirradiated spores of *Bacillus coagulans*. Ph.D. Dissertation, Purdue University, Lafayette, Indiana.

KUHN, G. 1960. Radiation protection of bacterial vegetative cells and spores. Ph.D. Dissertation, Purdue University, Lafayette, Indiana.

LEA, D. E. 1955. Actions of Radiations on Living Cells. Second Ed. Cambridge University Press, Cambridge, England.

MEISSEL, M. N. 1955. On biological effects of ionizing radiations on microorganisms. International Conference on the Peaceful Uses of Atomic Energy. Geneva. A/Conf–8/P/690 U.S.S.R.

MORGAN, B. H., and REED, J. M. 1954. Resistance of bacterial spores to gamma radiation. Food Research *19*, 357–366.

MULLER, H. J. 1928. The effects of X-radiation on genes and chromosomes Science *67*, 82–84.

POLLARD, E. 1959. Radiation inactivation of enzymes, nucleic acids, and phage particles. Rev. Mod. Phys. *31*, No. 2, 273–282.

PORTER, J. R. 1948. Bacterial Chemistry and Physiology. John Wiley and Sons, Inc., New York.

PROCTOR, B. E., and GOLDBLITH, S. A. 1952. Side effects in sterilization of food and drugs by ionizing radiations. Nucleonics *10* No. 4, 64–65.

RAHN, O. 1945. Death of irradiation. Bact. Rev. *9*, 8–14.

RAYMAN, M. M., and BYRNE, A. F. 1957. Action of ionizing radiations on micro-organisms. Radiation Preservation of Foods. U. S. Govt. Printing Office, Washington, D. C. p. 209.

STAPLETON, G. E., SBARRA, A. J., and HOLLAENDER, A. 1954. Nutritional aspects of bacterial recovery from ionizing radiations. J. Bact. *70*, 7–14.

TOBIAS, C. O. 1959. Effects of radiations on populations of cells and multicellular organisms. Rev. Mod. Phys. *31*, 290–297.

WAINWRIGHT, S. D., and NEVILL, A. 1955. Some effects of post-irradiation treatment with metabolic inhibitors and nutrients upon X-irradiated spores of Streptomyces. J. of Bact. *70*, 547–555.

WILLIAMS, N. J., and KEMPE, L. L. 1957. The effect of catalase on the lethality of cobalt-60 gamma radiation for certain anaerobic bacterial spores. Applied Microbiology *5*, 336–338.

WOOD, T. H. 1959. Some aspects of cellular radiobiology. Rev. Mod. Phys. *31*, No. 2, 283–289.

ZIRKLE, R. E. 1959. General features of radiobiological actions. Rev. Mod. Phys. *31*, No. 2, 269–273.

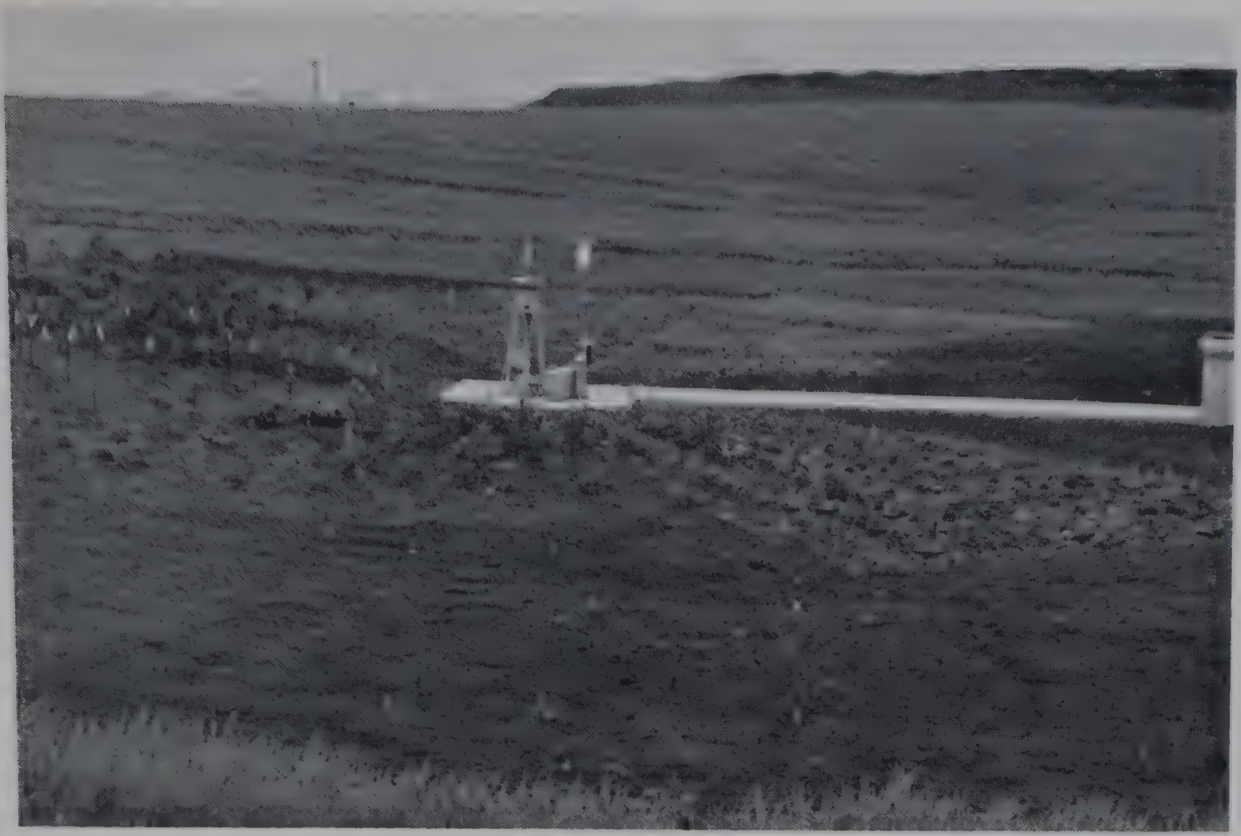
CHAPTER 13

Radiation Effects on Plants

While single-celled organisms are seen to be guided internally from the cell nucleus, multicellular organisms have differentiated and specialized cells which, in higher plants, are also susceptible to influences external to the cell in the form of hormones, permitting the cell to participate in the greater organization of higher life.

There is a linkage between a plant and its environment, with broad patterns of circulation or recycling of matter and energy. There is a complementarity of structure and function of the organism and its environment; plants vary widely in form and adaptability. Plant life on earth has not varied substantially in at least the past million years, indicated from fossils and excavations in very ancient caves. Plants living on the surface of the earth have always existed in at least a low level radiation field, and plants contain radioactive elements in greater or lesser degree depending on the soil from which they draw their nutrients. The levels of radiation encountered in nature apparently have insignificant effects except perhaps in causing occasional mutations, some of which no doubt are useful, the others disappearing from the life scene. However, the exposure of plants to more intense irradiation has dramatic consequences, and the more complex the organism, the more sensitive it is. The elimination of one key biosynthetic step by radiation in an integrated metabolic system has notable effects. A plant is a large system of complex molecules in delicate balance, and there are very few compounds that have ever been identified which have no recognized, useful function. And, there are alternate metabolic pathways occasionally available to some extent through adaptive enzyme systems. However, higher plants are unable to tolerate megarad radiation exposures which bacteria survive. Further, the measurable effects of a radiation treatment are most gross in the higher plant, and it is not always possible to trace the measured effects back to a sub-microscopic mechanism. Plant responses in radiation fields are morphological and physiological changes, mutations, and death.

A number of low level, cobalt-60 installations have been built in many countries to study plant responses to radiation. Fig. 72 shows such a facility located at Rise, Denmark. Essentially, these fields have a radiation source which is lowered into the ground to permit personnel access to the area. Plants and/or seeds are positioned radially from the source.



Courtesy of Danish Atomic Energy Commission

FIG. 72. COBALT-60 IRRADIATION FIELD

Plants positioned radially from ground zero (Rise, Denmark)

which subsequently is raised to expose the plants, the dose absorbed decreasing as the plant position becomes more removed from ground zero. A variety of interesting results have been found in using such facilities. In addition, plants are irradiated by exposure in reactors and reactor beams, particle accelerator beams, X-rays from generators, and to radioisotope emissions.

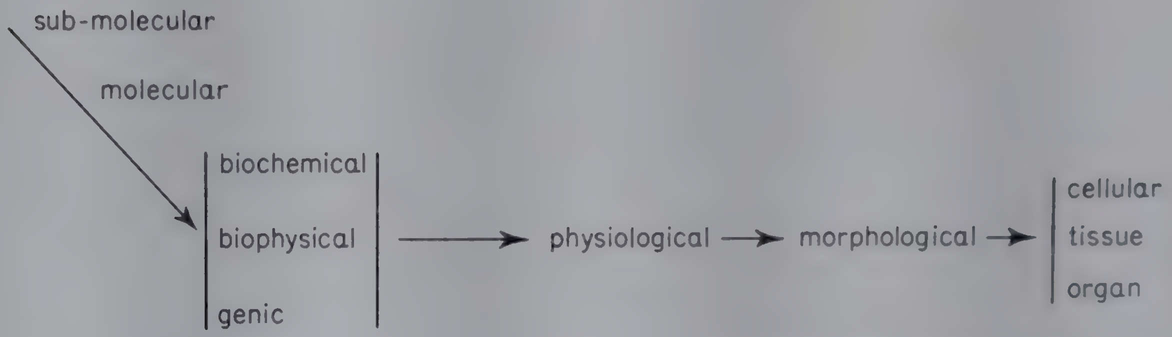
Sequence of Radiation Damage to Plants

The sequence of events which occur in irradiated plants is outlined in Fig. 73, the extent of the changes being mainly related to the dose absorbed. The interaction of radiation with the order and sequence of events embraced in higher plant growth and reproduction are most complex, yet some degree of understanding is possible, based on the level of present knowledge (Gordon 1957, Gray 1954).

In studying the responses of plants in radiation fields, it should be useful to review in broad strokes the growth and reproduction of plants in general, and then to undertake to explore the biological expression of radiation effects in terms of the physiological, morphological, and genetical responses.

Growth of a Plant

The growth of a plant is considered an irreversible process in which there is an increase in size, usually accompanied by an increase in weight and the amount of protoplasm. Growth is a quantitative matter concerned with increasing the amount of the organism. The process of development in a plant involves its changing shape, form, degree of differentiation, and state of complexity. Development is seen as a qualitative concept referring to changes in the nature of the growth made. The cycle of growth and development may be considered as follows: When a mature seed is placed in a suitable environment, germination occurs with the production of a seedling. The seedling in turn yields a mature plant. This process is the vegetative growth of the plant which is contrasted with the reproductive development which follows. The reproductive processes commence when a plant has attained a certain size, age, or a minimum number of leaves (Leopold 1955). The reproductive process of a plant first occurs in floral primordia and is first demonstrated in flowering, followed by fertilization, and the subsequent growth of the fruit which finally yields a developed seed. With the yielding of the seed, the cycle of growth and plant development is complete.



Courtesy of and adapted from Gordon

FIG. 73. BIOLOGICAL EXPRESSION OF ABSORBED HIGH ENERGY RADIATIONS

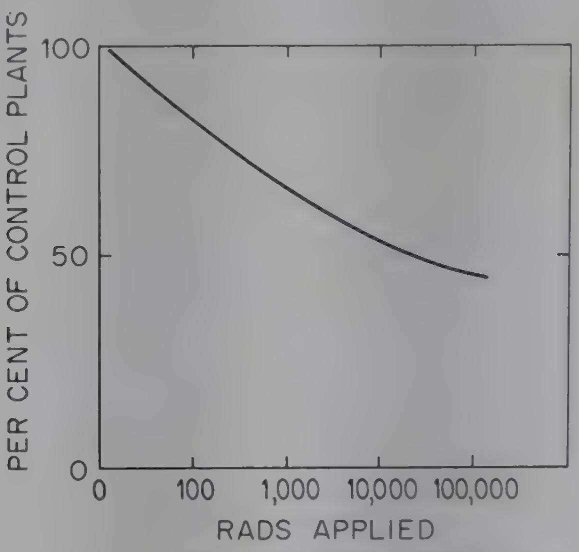
Zones of Growth.—The growth of a plant is concentrated in specific and characteristic zones. Generally speaking, the process of growth is initiated in regions of the plant in which cell division takes place, known as meristems. Primary growth is responsible for increasing the length and the production of branches and other lateral parts such as leaves and flowers. Secondary growth is responsible for increasing the diameter of plants. The process of cell division occurs in the meristematic regions, the process being discussed in Chapter 11. Cell enlargement is the most obvious phase of plant growth. This increase in size is caused primarily by the absorption of water, because of which the plant cell expands. When the cell wall is sufficiently plastic yet is irreversibly stretched, then true cell elongation has occurred. Elongation is generally accompanied by the creation of more cell wall. New cell wall development keeps abreast of elongation so that walls remain comparatively constant in thickness during growth. Cell enlargement is not only a passive increase in cell volume, but is also a synthetic process in which protoplasm formation has participated. Following or accompanying cell enlargement in plants there is a differentiation of cells which involves several kinds of anatomical and physiological specializations. Cells become specialized with respect to size and shape, or nature and extent of secondary wall development, or even with respect to their protoplasm. Differentiation is the transformation of apparently identical cells produced in the meristem into a variety of specialized cells. The manner in which these differences arise in genetically identical material is unknown. It is recognized that the growth of a plant is regulated not only by the environment or the minerals taken from the soil by the roots, or by the carbohydrates synthesized in the leaves, but also by a system of chemical entities which have specific roles as agents in synchronizing the growth of one part with the growth of the whole. These chemical compounds are synthesized in one tissue or organ of a plant and then are translocated to other sites where they are capable of producing special growth effects. These materials are called the plant hormones.

Growth Hormones.—Plant growth is dependent on the environment, on mineral uptake, on photosynthesis, and on the plant hormone systems. In the plant the principle of hormone regulation has been highly evolved. The integration of activities in the various parts of a plant is achieved by the synthesis, transportation, and utilization of these chemical messenger substances—the plant growth hormones. The specific substances required for root growth, for example, are produced in the leaves. Root growth is therefore in part regulated by the amount of root growth substances produced in the leaves then translocated to the root. The growth of young leaves depends on certain leaf growth substances which are produced in mature leaves. The growth of the stem depends in part on the growth hormones of the auxin type, which are produced in the apical bud and young leaves, auxin being transported to the growing regions of the stem. Auxins are involved in stem growth, root growth, in lateral bud initiation, abscission of the leaves, fruit growth, abscission of fruit from the plant, and in more than 20 different physiological activities in a plant. Auxins appear to be master hormones exercising control over many plant processes and perhaps even over other plant hormones. There is suggestion of a hormone which controls flowering (Leopold 1955).

The process of sexual reproduction in higher plants is initiated at the time of flower formation. With the initiation of floral primordia, the growth of a plant is altered and a developmental step occurs. Plants must make a certain minimum vegetative growth before they attain the opportunity of flowering. The initiation of flowering is but the first stage in reproductive development. It is followed by growth of the flower, pollination, fertilization, growth of the fruit, and the maturation of the ovary or seed.

Plant Responses to Radiation

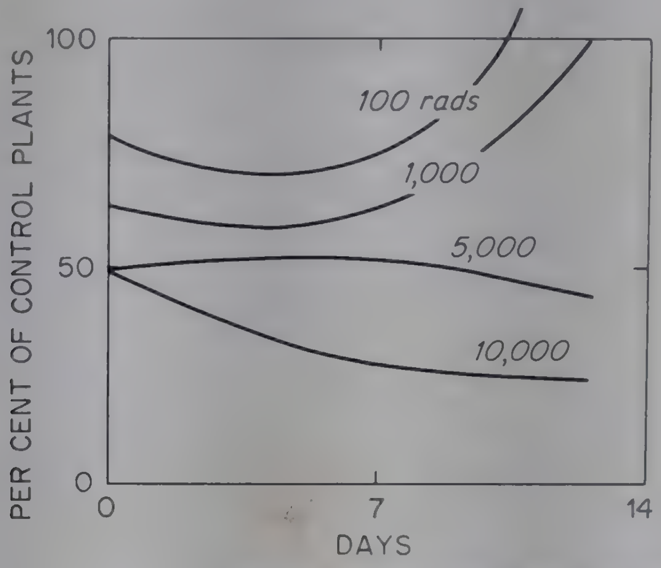
While the study of plant responses to ionizing radiation is a relatively new area of investigation, some biological effects are already becoming understood. Initially there is a change at the sub-molecular level of organization which, if not reversed within a short period of time, results in altered physical and chemical patterns of activity or in a modification of the genetic structures and relationships. These give rise to physiological changes ranging from minor alterations in form, to death of the organism, which might be defined as the inability of cells to reproduce at any organizational level in the plant. Classical studies (Gordon 1957) undertaken with plants irradiated even at 100 rad levels find an immediate reduction in the concentration of auxin present (Fig. 74). Many of the responses by plants in an ionizing radiation field have been attributed to the destruction of the growth hormone substances. At this level of irradiation, the actual energy absorbed by the plant is small indeed. Yet it is enough to cause direct chemical change in a few



Courtesy of and adapted from Gordon

FIG. 74. CHANGES IN FREE AUXIN LEVEL IMMEDIATELY FOLLOWING X-IRRADIATION OF WHOLE PLANT SHOOTS OR SEGMENTS

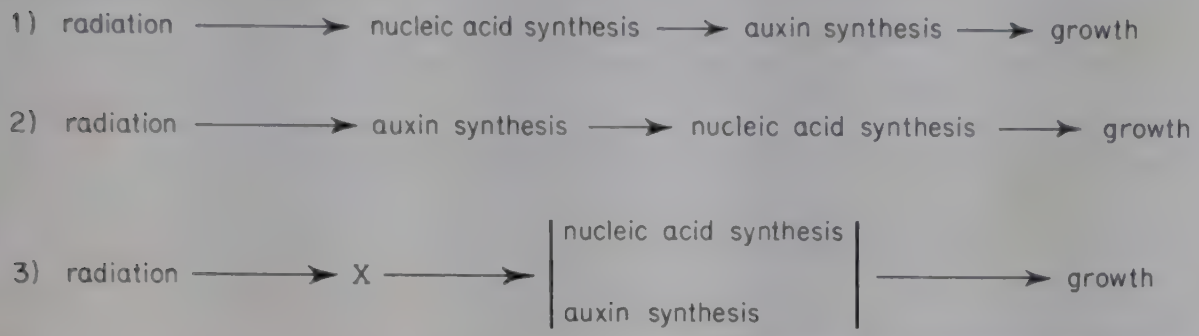
hundred molecules per cubic micron of tissue, and this happens to be the concentration of the free growth hormone present. Gordon has reasoned that irradiation leads to at least two possibilities: (1) The energy absorbed by the plant tissue is dissipated preferentially in auxin destruction; or (2) the energy is absorbed in activated molecules or molecular relationships that are responsible for the turnover of many auxin molecules. Furthermore there must be a mechanism that multiplies the effect originally due to the absorbed energy. The decrease in auxin concentration would thus be an indirect result of the destruction of a more primary radiation induced event. It appears from studies on the efficiency of auxin destruction in pure solutions and in tissues with ionizing radiation that the order of destruction is actually quite low, and it is probable that auxin in the plant is neither particularly radiosensitive nor preferentially inactivated. Therefore, it appears that the damage is caused by some other mechanism, perhaps indicating an enzyme whose damage would affect the turnover of many molecules. Newly formed shoots have active systems for auxin metabolism, as the growth hormone is being constantly synthesized in young buds. At the same time, hormones are being depleted by growth and by enzymatic and non-enzymatic systems for auxin inactivation. In those plants in which radiation does not decrease the auxin concentration and the plants are unable to synthesize auxin, such tissue shows lower auxin depletion rates. It is assumed that "free" growth hormones are present in a dynamic pool maintained as a steady state system by the concomitant biosynthetic and depletion reactions. If biosynthesis were preferentially and immediately radiosensitive, the rate of auxin formation would indeed be decreased or terminated. The level of free auxin in the pool would drop, the magnitude and rapidity of the decrease would be a function of the turnover rate of auxin. Conversely, the same result would be obtained if the rate of auxin destruction were preferentially accelerated by irradiation. It is currently thought that the auxin level following the irradiation of a plant is depressed because a rapid curtailment in auxin biosynthesis occurs rather than an accelerated auxin metabolism. When following the rate of enzymatic conversion of tryptophan to indoleacetic acid which occurs in the plant, doses as low as 10 rads result in approximately 10 per cent inhibition of this enzyme system, with inhibition increasing exponentially with increasing dose. The precursor of auxin increases in inverse proportion to indoleacetic acid as the dose is increased. The apparent pile up of the aldehyde suggests that a radiation block occurs in the enzyme system that oxidizes indoleacetaldehyde to indoleacetic acid. The response of the auxin forming system some two weeks after irradiation may find a pattern of recovery in biosynthetic ability. Complete or virtually complete recovery may occur in two weeks with doses below 1,000 rads, partial recovery with 2,000 rads, and none at 5,000 rads or above (Fig. 75). From physiological, morphological, and biochemical investigations, it has been shown that a relatively high radiation sensitivity of auxin biosynthesis exists (Gordon 1957). Other studies undertaken to estimate the enzyme destruction per unit of ionization show ionic yields in the region of unity. This is equivalent to saying that the energy absorbed is to a large degree diverted to the inactivation of auxin-forming enzymes, disregarding the molecular excess of other cellular chemical species. This enzyme appears as a relatively enormous radiation target.



Courtesy of and adapted from Gordon

FIG. 75. RELATIVE FREE AUXIN LEVELS OF PLANTS FOLLOWING IRRADIATION

In considering radiation damage, it is necessary to differentiate between the immediate primary effects of radiation on the plant and the changes that arise from disruption of key cellular and metabolic processes. It appears that there are threshold levels of molecular disorganization required before cellular damage can be perceived. The synthesis of nucleic acids is inhibited by irradiation and this can be identified shortly after an organism is irradiated (Gordon 1957). The synthesis of DNA occurs in meristematic tissues. Cells which have been irradiated at the mitotic stage do not elaborate DNA and the mitotic activity is delayed. The degree of inhibition is dose dependent, effects from as little as 35 rads being detectable. Gray (1954) has indicated that the pattern of inhibition and recovery of cells upon irradiation parallels almost identically the inhibition and recovery of mitotic activity in meristems. It appears that the inhibition of DNA synthesis causes the mitotic inhibition. Since both auxin and DNA are required for cellular multiplication, radiation damage to the growth of a plant may be related to both auxin and DNA biogenesis. The relationship which has been postulated is shown in Fig. 73. DNA is synthesized in the nucleus, while auxin is synthesized in the cytoplasm. Elongating cells are highly resistant to radiation damage. Since these cells do not synthesize auxin, but evidently do synthesize DNA, possibility 2 in Fig. 76 appears the most probable.


1) radiation → nucleic acid synthesis → auxin synthesis → growth
2) radiation → auxin synthesis → nucleic acid synthesis → growth
3) radiation → X → nucleic acid synthesis / auxin synthesis → growth

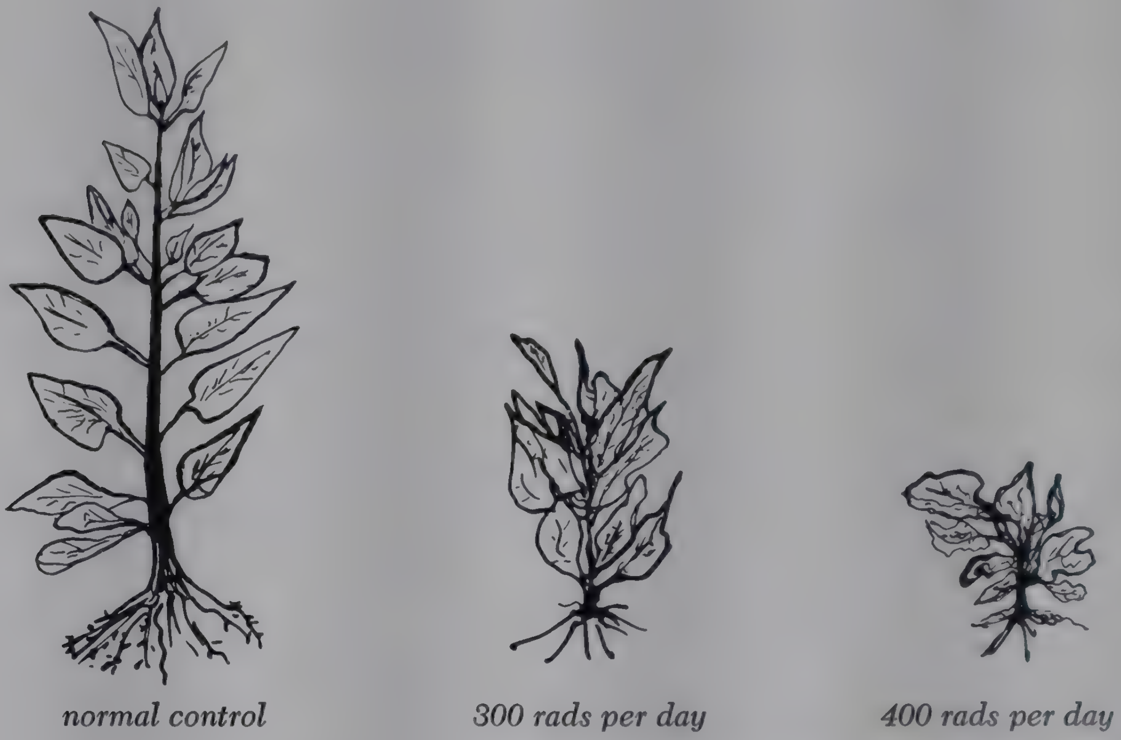
Courtesy of and adapted from Gordon

FIG. 76. POSSIBLE LOCI OF BIOCHEMICAL DAMAGE IN THE INHIBITION OF PLANT GROWTH BY IONIZING RADIATIONS

It is fascinating to consider how such a small amount of absorbed energy in the form of ionizing radiation can trigger a sequence of events that constitute the radiation syndrome. This area is under intensive investigation in many laboratories of the world.

Influence of Radiation on Chromosomes.—Ionizing radiation often causes chromosome aberrations; three general classes of aberrations are found depending on the stage of nuclear development at the time of irradiation (Bloom 1959). When irradiated at the resting stage, chromosome aberrations result. Chromosomes at this stage respond to radiation as if they were single strands. At the metaphase and anaphase, the induced aberrations involve both sister chromatids. The chromosome aberrations include deletions, dicentric chromosomes, and ringed chromosomes. Inversions and translocations also are produced (see Fig. 60).

The radiation at the prophase stage results in chromatid aberrations. At this stage the chromosomes have divided into sister chromatids, and the radiation may break both or either one at any given locus. Irradiation at very late prophase produces half chromatid or sub chromatid aberrations. The origin and appearance of typical chromosome, chromatid, and half chromatid aberrations are summarized by Sax. It is well established that cell division is inhibited by ionizing radiation, as has been discussed. The retardation of mitosis increases with increased dose. This suppression of mitosis and its recovery can be shown with radiation doses as low as 15 rads. Chromosomes in plant cells show great variation in their sensitivity to ionizing radiation at different periods in the nuclear cycle of mitosis and meiosis. The resting stage is most resistant to irradiation as measured by the frequency of chromosome aberrations. The relation between dosage and the frequency of the various types of radiation-induced chromatid and chromosome aberrations has provided information regarding the mechanism of aberration formation (Sax 1957). The fact that simple deletions have a linear relation to dosage indicates that these aberrations are the result of a single reaction or hit. The more complex dicentric ring and exchange aberrations tend to increase as the square of the dosage applied indicating that perhaps they are largely the result of two independent events or hits. Chromosome dot deletions appear to be predominantly the result of single hits. While the frequency of one-hit aberrations induced by a given dose of radiation is not dependent on radiation intensity, the frequency of occurrence of two hit aberrations appears dependent upon the intensity of the radiation. The radiation at high intensities produces more ring formation in chromosomes than does the same dose at low intensities. Chromosome aberrations produced by a given dose of radiation depend on the type of radiation, neutrons inflicting more aberrations for a comparable dose than gamma rays.



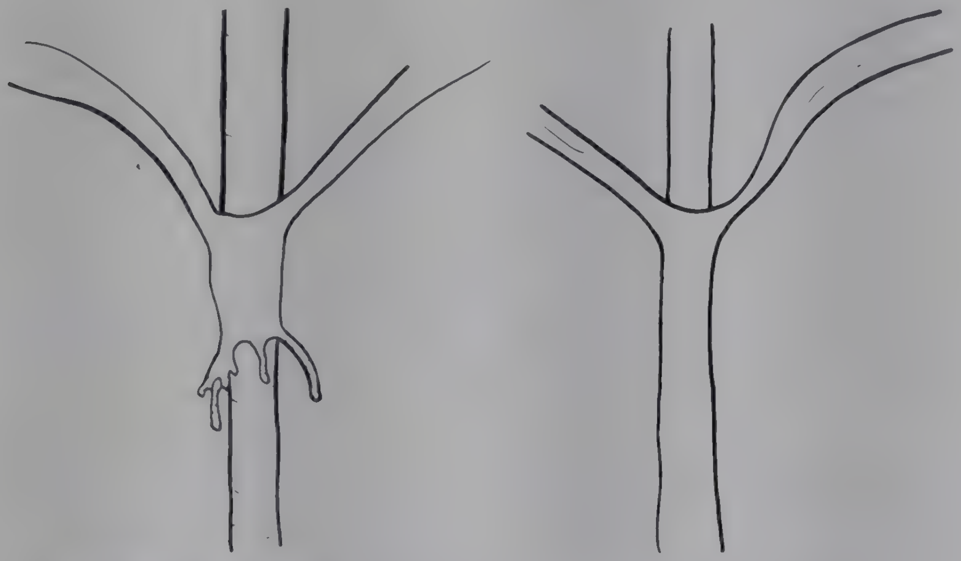
Courtesy of and adapted from Gunkel

FIG. 77. GROWTH OF PLANT INVERSELY PROPORTIONAL TO THE DOSE RATE—ABOVE THRESHOLD VALUE

Plants exposed at rates shown

Further, low energy neutrons are more efficient in producing chromosome aberrations than are high energy neutrons. Irradiation with alpha rays or neutrons shows a linear relationship between dosage and frequency of two-hit aberrations. The effects produced by gamma or X-rays, tend to increase in proportion to the square of the dose. The above differences apparently are attributable to the higher ionization density with alpha particles for example. It should be pointed out that the frequency of chromosomal aberrations induced by a given dose of irradiation is dependent upon the nuclear stage being irradiated, and the effect can be modified by varying the temperature of the cells during irradiation, by varying the dose intensity, by irradiating the cells in the presence or absence of oxygen, and by the treatment of cells with various chemical agents.

Morphological Changes in Plants Induced by Irradiation.—The modification in the growth and development of components of a plant detectable morphologically is well established. Gunkel (1957) has summarized the general morphological responses of plants to ionizing radiation as follows: To begin with, the results of studies with one species or variety of plants do not necessarily apply to others. Different types of responses are found in different plants and even different stages of development in the same plant. Secondly, while the responses observed are abnormal, they are not particularly new and may have been found in any study of monstrosities in structure which occur, due to one influence or



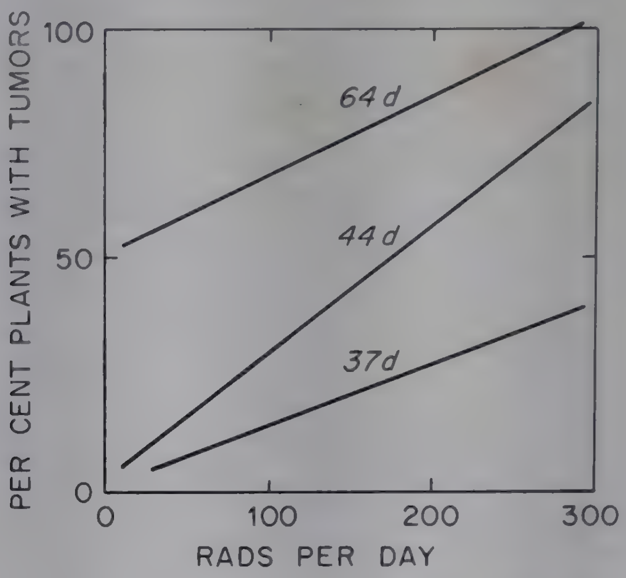
30 days after receiving dose of 3,000 rads

normal control

Courtesy of and adapted from Gunkel

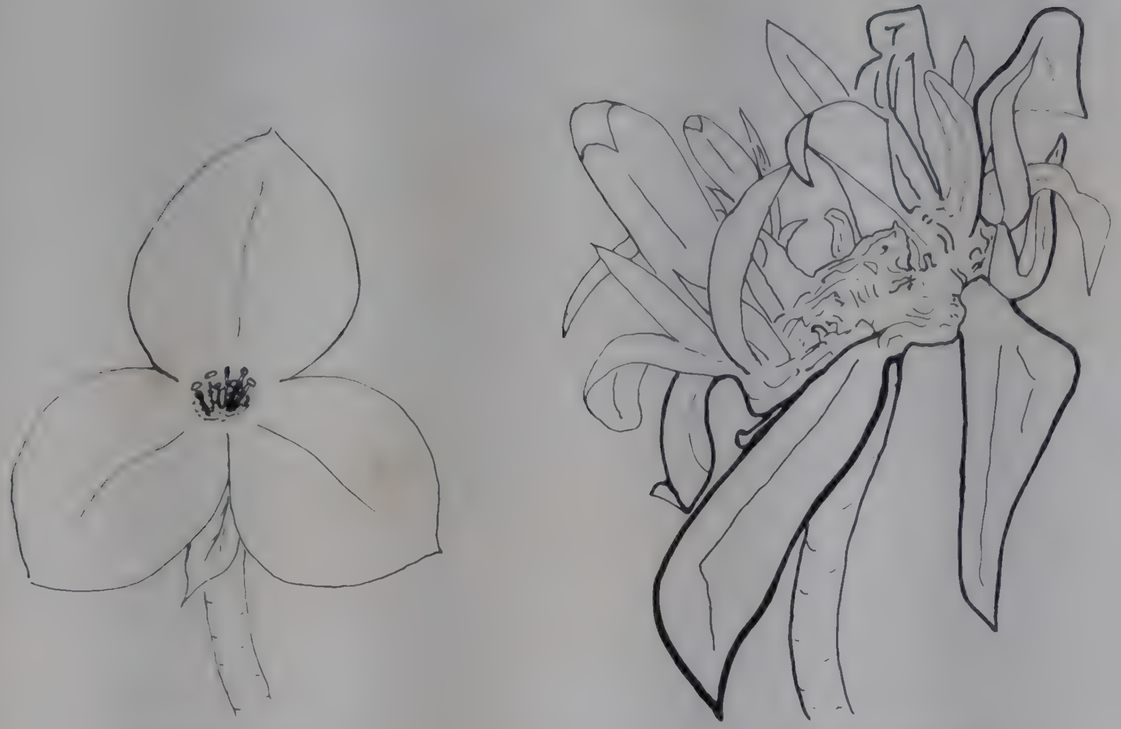
Fig. 78. Stem Swelling and Adventitious Root Production in *Xanthium* Stems

Base stem fails to enlarge following the irradiation another, for example weed killing agents. Radiation appears to speed up or accentuate events that are known in nature. Such information is particularly useful in the analysis of developmental changes induced by radiation, particularly where the events in growth are not observed easily because of the time factor. The morphological responses induced by radiation are the expression of the genetic and physiological factors previously discussed. On exposure to ionizing radiation, a particular plant may die, may have inhibited or even stimulated growth patterns, may demonstrate accelerated differentiation of cells, or even demonstrate morphogenic abnormalities. When stems are irradiated, they may be stimulated in dwarfing characteristics (Fig. 77), and have excessive branching. Some of the characteristic manifestations are shown schematically in Fig. 78. Irradiated stems may show fasciation, dichotomy, local stem swellings, increased amounts of vascular tissue, the formation of adventitious meristems, or tumors (Fig. 79). The effects of radiation on flowers leads to stimulation in height of floral stocks and abnormal thickness in fasciation, or irradiated plants may show delayed and reduced flowering or premature and increased flowering, color changes, a high degree of sterility, abscission, modifications in form and number of flowers, especially in petals and stamens, and the development of vegetative growth in floral positions. Some of these effects are shown schematically in Figs. 80–82. Fruit growth may be inhibited or stimulated depending on the dose rate of radiation applied. As shown in Fig. 78, the formation of adventitious roots is often associated with localized stem



Courtesy of and adapted from Gunkel

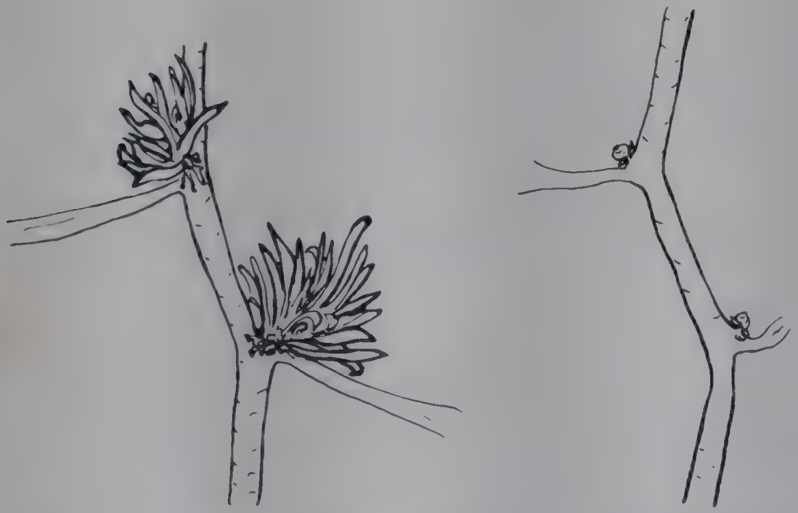
FIG. 79. RELATION BETWEEN THE DOSE RATE APPLIED PER DAY AND THE PER CENT TUMOROUS PLANTS AFTER VARIOUS PERIODS OF IRRADIATION



normal control *35 rads per day*

Courtesy of and adapted from Gunkel

Fig. 80. Vegetative Growth Occurs in Normal Floral Position Following Irradiation. When Removed from Gamma Field, and Allowed to Recover, Fused Shoots Rise in Floral Bud

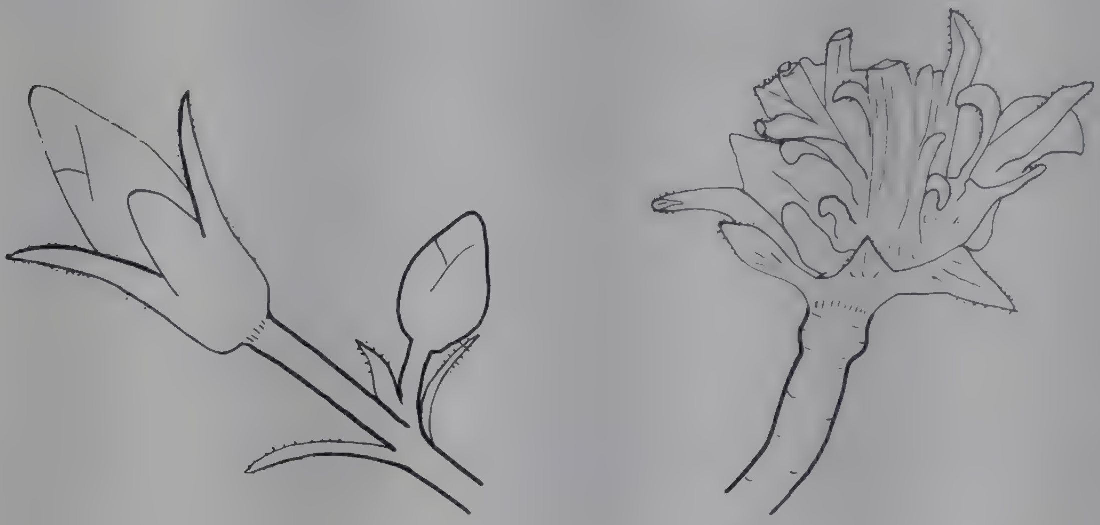


37 rads per day for 90 days *normal control*

Courtesy of and adapted from Gunkel

Fig. 81. Modified Auxillary Bud Expression Following Irradiation of *Trandescantia paludoso*, Adventitious Buds Formed Producing Irregular Leaves and Multiple Shoots (37 Rads/Day, for 90 Days)

swellings. With high irradiation doses root growth is generally inhibited. It is to be recalled that a root is a very conservative structure and is well shielded from radiation by its favorable location in the soil and its ultra low level of radioisotopes in ordinary circumstances. The leaves of irradiated plants may show changes in color, form, and texture, after irradiation. Some tumorous growths have been reported as a response to local injury. Gunkel (1957) has summarized the response to include mosaic-like color changes or an overall color change usually accompanied by increased pubescence, premature abscission, irregular blade development, which would include a range of distortions. The failure of the leaf blade to develop adequately is demonstrated schematically in Fig. 83. It is noteworthy that these malformations are also found in unirradiated plants.



control

625 rads per day for several months

Courtesy of and adapted from Gunkel

FIG. 82. FLORAL ABNORMALITY IN *Oxalis* DUE TO IRRADIATION OF FLORAL BUD

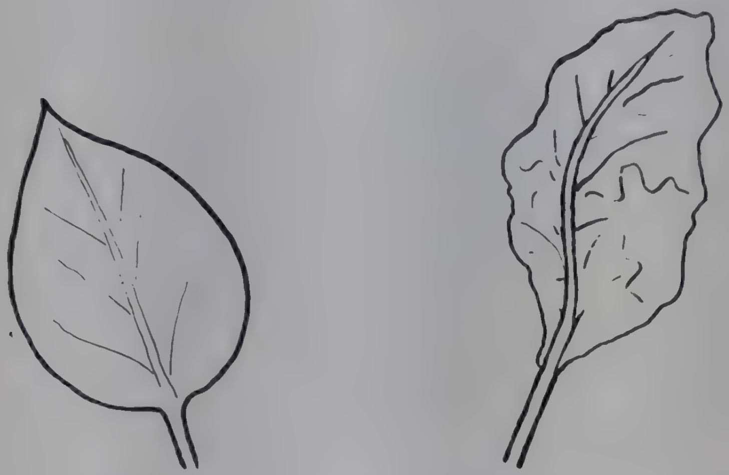
Petal-like structures develop in normal floral position

Responses of a plant to ionizing radiation are complicated and diverse. The morphological effects demonstrated appeared to be secondary effects due to physiological imbalances in plants. Excellent reviews on the subject are available and are listed in references.

Useful Applications

That ionizing radiations have widespread influence on plant growth and development is well established. With the development of low intensity gamma radiation fields such as shown in Fig. 72, a number of useful applications have been found. Included in these have been the development of useful mutants which, when incorporated into standard varieties have yielded beneficial results (Gustaffson 1958). Another interesting result was uncovered by Sparrow (1957), involving the inhibition of sprouting in potato tubers when subjected to ionizing radiation. This has evolved into a most useful process, which has found profitable application (Brownell *et al.* 1957; Sparrow and Christensen 1954; Heiligman *et al.* 1958).

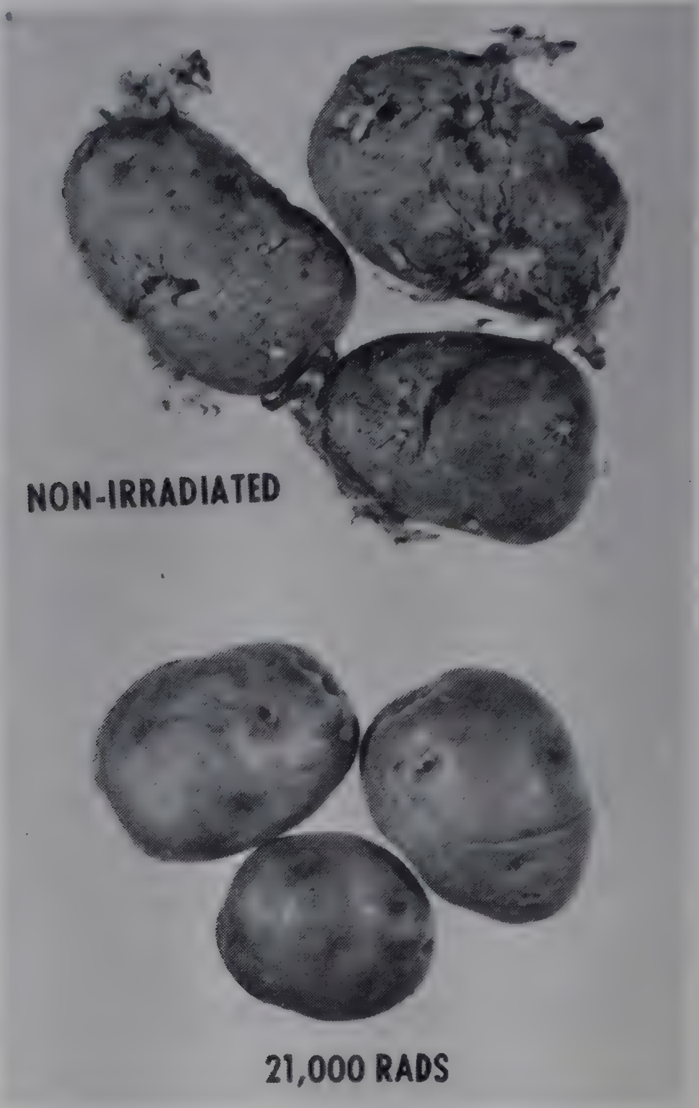
Interruption of Plant Growth with Ionizing Radiation.—When potato tubers are planted in soil which is then exposed to an intense radiation source, the tubers nearest to the source do not sprout or grow. When the potatoes are dug from the ground, it is observed that the tuber is indeed sound, the only apparent manifestation of the radiation treatment being the inhibition of sprouting. Because this is an important finding in radiation technology it will be discussed in some detail. The economic importance of the potato in the diets of mankind is well established. One of the principal losses on storage is due to sprouting of the tubers, resulting in losses in nutritive value, weight, and usefulness. The finding by Sparrow and Christensen (1954) that potato tubers exposed to gamma radiation from cobalt-60 would indeed not sprout might at first glance appear to be a negative result. In the use of such tubers for seed stock in new plantings it would also appear to be a disadvantage. However, the application of this treatment to the improved storage and distribution of potato tubers is equally apparent. Initial tests with potatoes showed that irradiation with dose ranges from 1,000 to 1,000,000 rad treatment caused notable


control *150 rads*

Courtesy of and adapted from Gunkel

FIG. 83. LEAF ABNORMALTIES RESULTING FROM CHRONIC RADIATION DAMAGE

Leaf becomes thickened and puckered, leaf blade irregular in outline (tobacco leaf)



Courtesy of Brownell

FIG. 84. TREATED AND CONTROL POTATOES AFTER ONE YEAR'S STORAGE AT 41°F.

deterioration in the potato quality. This is indicated in Fig. 84 from the work of Brownell and his associates (1957) at the University of Michigan. Early experiments revealed that factors other than sprout inhibition must be considered in order to establish a process for prolonging the storage life of potatoes. Varietal differences, the time of irradiation relative to harvest and dormancy, the curing of the tubers, the storage environment, and other factors as well as the radiation treatment, appear to play important roles. A large number of research projects have operated with the express purpose of yielding specific information necessary to establish successful applications of radiation for sprout inhibition and increased storage life of potatoes, onions, and root crops.

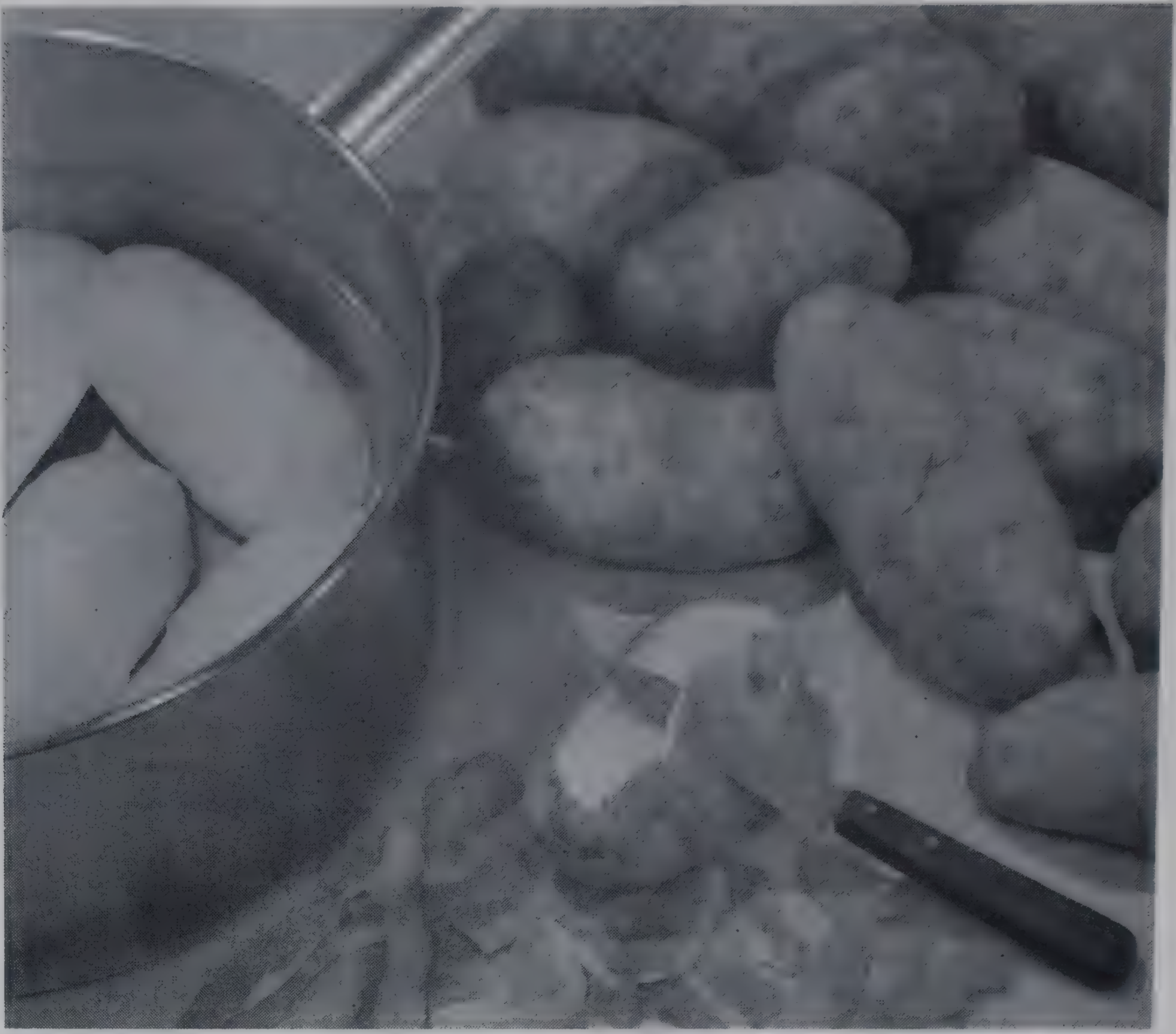
At 5,000 rads sprouting is drastically retarded in potato tubers and completely inhibited by 20,000 rads (Brownell *et al.* 1957). Weight losses during storage are markedly reduced at all levels of radiation in which sprouting was inhibited. After prolonged storage of tubers at 35° and 50°F. both the controls and the tubers treated with only 1,000 rads show severe softening, weight loss, and sprouting. As is now known and indicated above, doses of radiation in the 1,000 rad range did not permanently interfere with the growth process.

As will be discussed in a later chapter, it is important to establish whether the irradiation treatment produces any harmful effects to potential human consumers. These tests include long term (2-year) animal feeding studies. Late in the fall of 1955 several tons of Idaho Russet Burbanks, Maine Katahdin, and Maine Russet Burbank potatoes were irradiated for such a purpose. These potatoes were also intended for use in studies of other factors, such as the behavior of the irradiated potatoes under simulated commercial distribution conditions, processing characteristics of irradiated potatoes, and the economics of the irradiation process. Because of the large quantities of potatoes involved, the irradiation was done at the Material Test Reactor Site at Idaho Falls, Idaho, using gamma radiations from spent fuel rods. Each variety was divided into three lots, two for irradiation with treatments at two dose ranges, and one an unirradiated control group. In the low range, none of the tubers received less than 15,000 rads or more than 22,000 rads, while in the higher range, none received less than 30,000 rads or more than 44,000 rads. It was not possible to control dosage over a smaller range due to physical and mechanical problems existent at that time and with the technology available in 1955. After irradiation, the potatoes were shipped to Maine, where they were stored in commercial potato storage bins. After a short period of storage, decay had occurred in all lots with definite increases of rot in the treated lots over the non-irradiated controls. Since all of these potatoes had received a great deal more handling than they would have received in normal commercial practice, it was believed that the major contributions to the onset of rot was the excessive handling. The cause of the higher levels of rot in the treated lots could have been due to any one or combination of many factors, such as excessive dosage, failure of the tuber to recover from the radiation treatment, lack of wound healing due to an improper environment, or failure to form peraderm or to suberize. The tests on these potatoes were a failure, and animal feedings of such tubers were also negative. Biochemical studies on these tubers indicated that there was no significant difference between the lower level irradiated potatoes and non-irradiated controls. The factors studied were moisture content, specific gravity, total sugars, reducing and non-reducing sugars, ascorbic acid, starch, and respiration. One exception was found with the Maine Katadhin potatoes, in which the irradiated lot showed a higher total sugar content after the fourth month of storage and through the twelfth month.

Early investigations on the physiology of irradiated tubers indicated that the treatment affected the processes involved in periderm formation and suberization, and destroyed the ability of the potatoes to sprout, or evidence growth. Any impairment of these processes must be considered important in the successful storage of root crops since these are the mechanisms which aid in wound healing and in preventing excessive losses and in preventing decay. Radiation treatments between 32,000 and 80,000 rads affect the wound-healing ability and do not increase the tuber's resistance to decay. Tubers treated with 12,000 rads did not sprout and if held under optimum conditions, cool temperatures and high relative humidities, develop periderm. Studying several varieties of potatoes and several environmental conditions, suberization is found to be delayed following irradiation but the suberized layer of the treated tubers ultimately thickens, as does the controls'. Periderm formation is altered at all levels of radiation from 1,000 upwards. Treatment with 5,000 rads completely inhibited sprouting in certain potato tubers, whereas some sprouting was found in other varieties with 10,000 rads. At 10,000 rads, sprouting was inhibited in the Russet Burbank variety. In studies on weight losses, the losses in tubers for a given storage environment decreased with increasing dosage up to 20,000. In general, the data suggested that a low dosage, the minimum for sprout inhibition, would be more effective in controlling weight loss during storage than higher dosage. The preliminary data then demonstrated that the radiation treatment must be confined to the lowest useful dose which inhibits sprouting. Using whole tubers, the respiration rate, measurement of the carbon dioxide evolved, when compared to non-irradiated controls, decreased immediately after irradiation and within a few hours increased to one and a half times that of the untreated controls. Oxygen uptake by potato slices increases as a result of irradiation. This increase occurs immediately and continues for some fifteen days. Oxygen consumption decreases after this time following the initial rise. The carbon dioxide production likewise remains high during this period. After several weeks the rate of respiration returns to that found with unirradiated controls, and from about the seventh week on, potato tubers respire at approximately the same rate as controls. Potato tubers which receive dosages of 50,000 and higher rads respire at higher rates than controls for long periods of time. Other studies showed that lots which received low level irradiation respired equally with untreated controls. No difference in acceptance of potatoes by consumers can be detected between the treated lots and the untreated controls at the low level irradiation level. One difficulty experienced with irradiated potatoes receiving 15,000 rads or more is that after cooking there is a darkening of the flesh when exposed to air. This is variety-specific. The radiation also seems to have the effect of increasing the intensity of the black spot which is known to be linked with certain varieties of potatoes. Studies on the control of post-harvest diseases of potatoes indicate that tubers treated with radiation doses of over 50,000 rads progressively become more susceptible to injury and hence attack by fungi. However, many potato pathogens such as *Fusarium* and bacterial "soft rot" inoculated into potatoes and irradiated at 50,000 rads are killed.

In studies on the susceptibility of potato tubers to rot, it is found that rot gradually increases at increasing intensity of radiation applied. Non-artificially wounded and non-artificially inoculated potatoes develop little storage rot at levels of radiation below 15,000 rads, with sprouting completely inhibited. Inoculated and wounded potatoes have increasing rot incidence with increasing radiation dose applied. The rate of rot progression through tuber tissues is more severe when the inoculation follows irradiation than when inoculation precedes irradiation, suggesting that the irradiation has some effect. No doubt it permits the more easy entrance of such organisms into the flesh.

While these initial studies were useful in yielding information on sprout inhibition and storage problems in potatoes, it remained to be demonstrated that the process was useful. In a second series of experiments begun in 1956, and terminated in 1959, sprouting was prevented by 5,000 rad applications to all varieties tested except the Red Pontiac, which required 7,500 rads, when tubers were stored at 41°F. When stored at 47°F., 7,500 rads was the lowest level of irradiation applied to tubers stored at this temperature which inhibited sprouting completely in all varieties. At 55°F., 7,500 rads inhibited sprouting in all varieties except Red Pontiac. This variety required 12,000 rads for complete inhibition. In all cases from a practical viewpoint, inhibition was accomplished between 5,000 and 10,000 rads. At 41°F. the weight loss of treated and control potatoes was rather similar up to 200 days after harvest. Beyond this time, sprouting was significantly decreased in the irradiated potatoes, which markedly diminished the rate of weight loss. At 41°F. little differences were found in the rate of weight loss during storage that could be attributed to pre- or post-irradiation curing of the tubers. At 55°F. storage, all potatoes generally showed a greater weight loss, but the unirradiated controls showed significantly greater losses. Decay of irradiated potatoes stored at 41°F. and 85 per cent relative humidity even after 600 days after harvest was negligible (Fig. 85). The higher the temperature of storage, the more rapid the advance of decay. After one



Courtesy of Workman

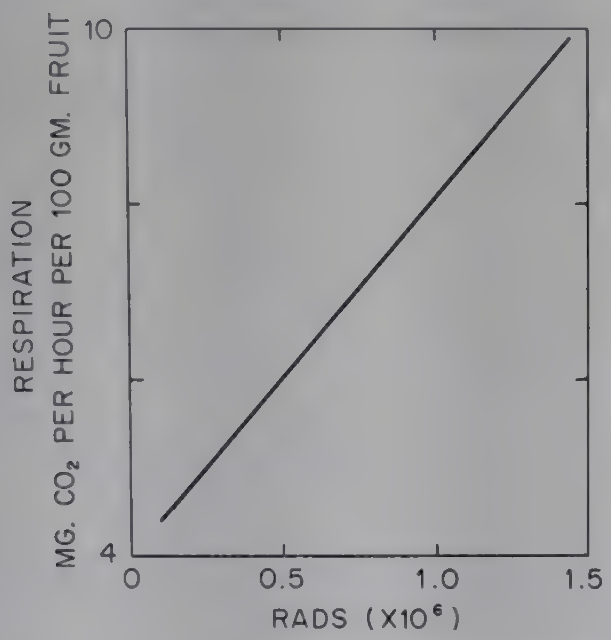
FIG. 85. TWO YEAR OLD IDAHO RUSSET BURBANK POTATOES AT 41°F., 85% R.H.

and a half years storage at 41°F. the Russet Burbank potato was found to lose the least weight and was excellent in appearance. Similar observations were made with other quality factors in potatoes. After one and a half years of storage after harvest, Russet Burbank potatoes were subjected to consumer evaluation, and the potatoes were found to be most acceptable. Potatoes stored for two years after harvest at 41°F., 85 per cent relative humidity, irradiated at 7,500 rads, evidenced some 10% weight loss. Sprouting was inhibited, and the potatoes were useful in cooking (Heiligman *et al.* 1958).

To be successful, it is important that good storage practices be used for the tubers, as well as application of the known technology in the handling of potatoes. Potatoes which are injured or under attack by rotting organisms, or otherwise of low quality, must be eliminated from consideration. If sound potato tubers are used and the above technology is applied, it has been found that potatoes may be retained in excellent culinary quality from one potato harvest period to the next, and the storage may be extended to two years if conditions warrant it. The magnitude of the opportunities is tremendous, and represents an important event in radiation technology.

In addition to the commercial possibilities of sprout inhibition of potatoes, it has been found by research workers in England that the application of radiation doses below 1,000 rads slightly retards the ability of the tuber to push forth sprouts, thereby enabling the distribution of potatoes for seed purposes in ordinary transportation systems. Potatoes are found to be retarded slightly, and when planted in the ground recover from the initial radiation treatment and proceed to produce useful plants and tubers. The rate of mutations found in such treated potato stock appears to be negligible, an occasional flower alteration having been noted.

Irradiation of Fruit.—The possibility of interfering with the metabolism of living plant organs has useful features in altering the maturation rates of fruits. For example, the normal respiratory pattern for ripening tomatoes finds an increase from a pre-climacteric low rate of carbon dioxide evolution at the mature green stage of development to a high climacteric at the point of the first development of red color. This is followed by decreasing rates as the tomato changes from a light pink to dark pink and eventually reaches a respiration rate comparable to the pre-climacteric low at the table or eating stage. Irradiation alters the respiratory activity, color development, pigment biogenesis, and the normal maturation process in general. The rate of respiration increases im-

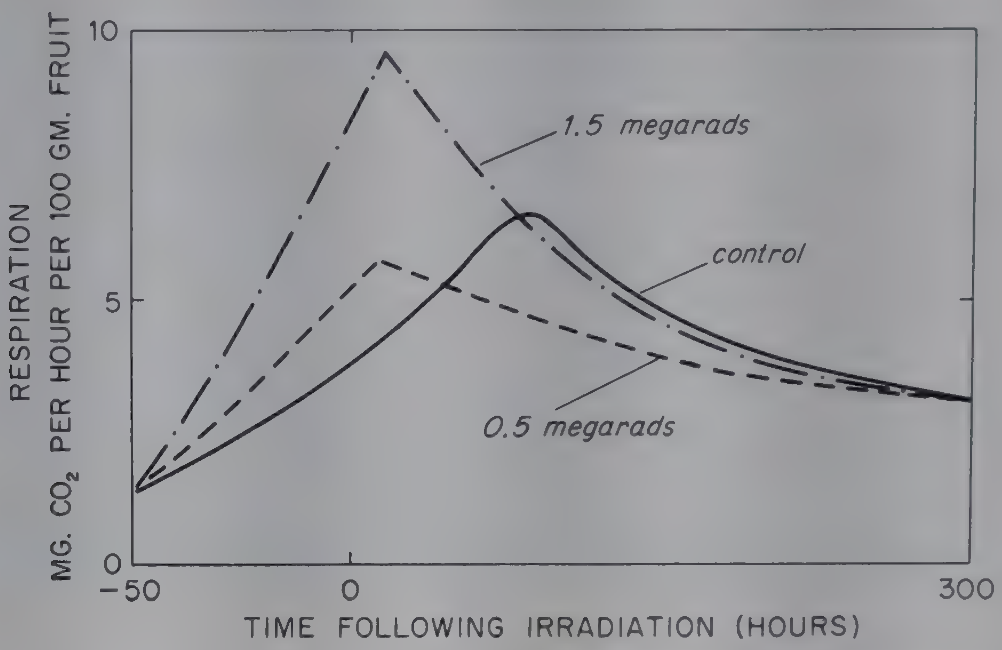


Courtesy of Burns and Desrosier

FIG. 86. RELATION OF DOSE APPLIED TO FRUITS AND THEIR RATES OF RESPIRATION (TOMATO)

mediately, proportional to the dose applied to the fruit, and the effect lessens with time. The effect on color pigment development at low doses is not of a permanent nature, and fruits usually recover.

Fruit immediately increases in respiratory activity proportional to the dose which is applied (Burns and Desrosier 1956). The effect with moderate doses is shown to persist in significant levels for at least 100 hours at the end of which time no statistical differences are found between the rates of carbon dioxide evolution of treated and control fruits. At 500,000 rads, tomatoes are apparently able to recover from the radiation exposure. Normal respiratory activities appear only incompletely disrupted. On the other hand, the dose necessary for the inactivation of typical bacterial and fungal fruit pathogens is bound to be much less than a minimum dose applied to these fruits, which would indicate that sterilization of fresh fruits can be effected with little disruption of respiratory mechanisms. The relation between the radiation dose applied and the change in the rate of respiration of tomato fruits is shown in Fig. 86. The rate of respiration of treated and control fruits in respect to time following irradiation is shown in Fig. 87.



Courtesy of Burns and Desrosier

FIG. 87. RESPIRATION VALUES FOR TOMATO FRUITS IRRADIATED AT TWO DOSES AT ZERO TIME

The ability to alter the maturation rate of fruits is of significance. An interesting series of studies undertaken in England shows that the ripening process of pears may be manipulated by the judicious use of radiation. The opportunities with many fruits appear widespread.

In the course of studies of the effects of ionizing radiations at doses greater than 200,000 rads upon the maturation of fruits, it is commonly observed that the treated fruits in storage exhibit susceptibility to fruit pathogens greater than controls (Table 32) (Burns and Desrosier 1956).

TABLE 32

INFECTION OF TOMATO FRUITS INOCULATED FOLLOWING IRRADIATION AT 1 × 10^6 RADS

Incubation Time (Hrs.)	Lesion Diameter, mm., Average Values	
	Control	Irradiated
48	6.6	10.5
120	9.7	13.6
360	30	35

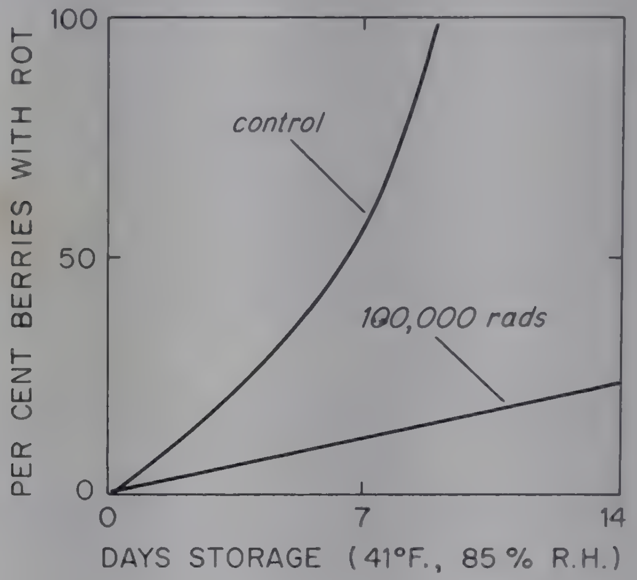
The spread of soft rot pathogens is greater in inoculated irradiated fruits than in inoculated controls. The increased susceptibility is explainable in terms of the pathological histology of the micro-organisms. Their movement through vegetative tissue is intercellular and it is preceded by the pectolytic enzymes associated with the organism which dissolves the pectic middle lamella between cells. These cells become loosened one from the other; protoplasmic membranes are ruptured which permits the exudation of cell sap into the intercellular spaces, where it provides the moisture and nourishment required for a rapid multiplication of the fungus and neighboring organisms.

The depolymerization of pectin with ionizing radiations has been previously noted, and should be assumed to be a major factor in predisposing irradiated fruits to this type of infection. The action of pectolytic enzymes conceivably should be enhanced as the pectic substrate in the middle lamella should have already suffered some degree of degradation due to the radiation treatment. It might be postulated that the stored carbohydrate material of the parasitized cell would also be in a more readily available form of nourishment for the fungus in view of the hydrolytic effect of irradiation also in degrading starch. It is seen that starch must first be hydrolyzed before being utilizable by organisms for energy.

However, in spite of this, surface sterilization is certainly effective at the rates from 100,000 to 200,000 rads. It would be expected that the surface contamination of organisms would be drastically reduced or completely eliminated if the area were treated with these doses. However, in the event that recontamination or incomplete surface sterility of fruit pathogens occurs, the resultant infection might be expected to proceed at accelerated rates once initiated. The problem of recontamination of fruit surfaces following irradiation fortunately is easily solved by suitable packaging and exposing specimens in sealed flexible films with suitable gas permeability characteristics for fruit respiration.

The irradiation of small fruits, such as strawberries, has led to interesting results. The fresh strawberry is highly perishable, and in the United

States the crop in California warrants attention. The crop in 1960 has a value of approximately $35,000,000. The fresh marketing of strawberries from California to New York markets is restricted by the ability to move the fresh fruit without decay from coast to coast by surface transportation. Transit requires five days. The life of a fresh strawberry in cold storage is approximately six days. Any interruption in transit has drastic results. As a consequence, from 12 to 25 per cent of the crop which is marketed in this manner suffers rot. This is a substantial economic loss. It has been found that the application of 100,000 rads to strawberry fruits sterilizes the surface sufficiently to permit simplified distribution techniques. Small baskets of strawberry fruits containing an inoculated berry in the center when irradiated and stored in cold storage show that after 15 days the inoculated berry has shriveled, and the infection of the other fruits which normally would occur does not happen. It is seen that low level irradiation of strawberry fruits, therefore, has significant economic import in at least doubling the useful distribution life of fresh strawberries, offers an opportunity for recovering the staggering economic loss rot inflicts, and may have marked importance in determining in which area of the country strawberries will be grown in the future. The incidence of rot in treated and control strawberries is shown in Fig. 88. Application to other small fruits appears to be most promising.



Courtesy of Maxsie

FIG. 88. RADIATION PASTEURIZATION OF STRAWBERRIES DECREASES LOSSES AND EXTENDS DISTRIBUTION LIFE

An improvement in the storage stability of sweet red cherries (usually a matter of days) has been found using ionizing radiation treatments.

Ionizing radiation treatment results in an improvement in the storage stability of sweet red cherries (usually a matter of days).

Cherries have been retained in excellent condition for 90 days, which is remarkable considering the highly perishable nature of these fruits. It is interesting to note that fruit irradiated at 100,000 rads or less, while being sterilized at least surface-wise, continues respiration, continues biogenesis, continues flavor development, and may yield fruits of greater consumer acceptance than untreated controls. The ability to degreen citrus commodities is also noteworthy.

Altering the Starch and Cellulose Content of Plant Structures.—That cellulose may be made water soluble when exposed to ionizing radiation has been discussed in an earlier chapter. The application of ionizing radiation to alter the structural components of edible plants appears most rewarding. Asparagus is notable in developing fiberous systems as it matures. Low level irradiation results in hydrolysis of such components resulting in more tender shoots. The applications of low levels of ionizing radiation to peas effects starch conversion to sugar. It is a common experience to find irradiated plant tissues sweeter to the taste than unirradiated controls. The irradiation of sawdust may also yield useful hydrolyzates. These possibilities have yet to be fully explored or developed.

Irradiation of Seed.—Vidal (1959) has found that the irradiation of seeds at low levels produces remarkable responses in the subsequent plant growth. As noted in plantings in cobalt-60 radiation fields, plant growth measured in height and dry weight matter at increasing distances from ground zero yields maximum plant responses which are greater than control plants which have not received the radiation treatment. Irradiation of many varieties of seeds has led to the conclusion that extremely low doses (less than 900 rads) increase the yield of the edible crop over controls. For example, the irradiation of radish seeds produces not only a doubling of the yield, but yields roots which are fleshy and tasteful, which is not common in too large radishes. The yield and the quality were both improved substantially by this treatment. Similar treatment of spinach caused doubling of each leaf. While this work is in its infancy, the opportunities for increasing the yield of edible plants from existing seeds are highly significant. It would be anticipated that such an irradiated seed would not form a suitable foundation stock for seed production. However, as is the case with hybrid corn, for example, seed production is isolated from crop production practices. It would appear that similar techniques will be required for irradiated seed used for crop production. When it is recalled that hybridization of plants yields approximately 15 per cent increases in crop yields, the returns from low level, inexpensive radiations certainly warrants careful attention.

Useful Plant Mutations.—Programs of study have evolved using mutated material in plant pathology, plant breeding, and entomology studies and in the fundamental evaluations in the field of genetics. The largest effort of this kind has been the work of Gustaffson in Sweden. Substantial plant radiation programs are also under way in the United States, sponsored largely by the Brookhaven National Laboratory. Initial work began with the irradiation of seeds and plant organs, seeking mutations. In 1949, a gamma radiation field was designed by Sparrow (1957) which allowed plant irradiation at any desired interval in the growth stage or cycle period. Such a facility was shown earlier. Substantial numbers of improved crop varieties have been obtained, some varieties resistant to disease organisms, others with desirable growth characteristics, such as stiffer straw, higher yield, and so forth. It has been learned that many factors govern the radiosensitivity of plants and it appears that eventually the plant breeder may be able to control some of the induced mutation processes in the direction of desired changes. A mildew-resistant barley mutant with high yield potential has been produced from irradiated seed in Austria. A barley mutant of equal interest has been found in Ireland with shorter straw than the parent and its kernel more round. Hybridization has been continued with this strain, attempting to place in it the desirable characteristics of other new varieties of high yield potential. A four-row mutant has been produced by cobalt-60 irradiation of a two-row fruit fly-resistant barley in Switzerland. It is hoped that the resistance will be retained in the mutant, and that the four rows will give a higher yield potential, features now being evaluated. The irradiation of graft units for various kinds of fruit has been studied in Yugoslavia but no positive results have accumulated to date. In Denmark, at Rise, seed irradiation work has also yielded a sterile male sugar beet.

The simple irradiation of a plant or a seed followed by its cultivation usually yields negative results (Gustaffson 1958). It is not to be anticipated that such a simple approach would yield highly useful, completely developed strains. However, the ability of radiation to induce mutation is real. Certain features are commonly seen in radiation induced mutations of plants which in themselves may not be useful, but which when combined with other known useful genetical qualities of other varieties, does indeed yield useful products. The effort is tedious and long range, which has discouraged many workers. Radiation then appears as a useful tool in plant breeding and in genetics. Beneficial results are beginning to appear. The manipulation of commonly known plants not only edible but ornamental in nature also has substantial value.

It is to be recalled that the genetical effects of ionizing radiation can be modified by changing the environment, e.g., by manipulating the temperature and moisture content of the irradiated material and by choosing various types and doses of ionizing radiation, all of which offer wide opportunities for the production of desirable mutations.

BIBLIOGRAPHY

ARNASON, T. J., PERSON, C. O., and NAYLOR, J. M. 1952. Radiation induced mutations in wheat and barley. Canadian J. Botany *30*, 743–754.

BLOOM, W. 1947. Histological changes following radiation exposures. Radiology *49*, 344–347.

BLOOM, W. 1959. Cellular responses. Rev. Mod. Phys. *31*, No. 1, 21–29.

BROWNELL, L. W., GUSTAFSON, F. G., NEHEMIAS, J. V., ISLEIB, D. R., and HOOKER, W. J. 1957. Storage properties of gamma irradiated potatoes. Food Technol. *11*, 306–312.

BURNS, E. E., and DESROSIER, N. W. 1950. Maturation changes in tomato fruits induced with ionizing radiations. Food Technol. *11*, 313–316.

CHANDHURI, K. L., and DAS, A. 1956. Effects of X-rays on the fertility of pollen grain. Science and Culture *21* No. 9 550–553.

GORDON, S. A. 1956. Auxin biosynthesis—a cytoplasmic locus of radiation damage. Progress in Radiobiology, Mitchell, J. S., Ed. Oliver and Boyd Publ. Co., London, England.

GORDON, S. A. 1957. The effects of ionizing radiation on plants: biochemical and physiological aspects. Quart. Rev. Biol. *32*, 3–13.

GRAY, L. H. 1954. Some characteristics of biological damage induced by ionizing radiations. Radiation Research *1*, 189–213.

GUNKEL, J. E. 1957. Effects of ionizing radiation on plants—morphological effects. Quart. Rev. of Biol. *32*, 41–56.

GUSTAFFSON, C. A. T. 1958. Radiation induced mutations in plants. International Meeting on the Application of Atomic Science in Agriculture and Food, E.P.A.—O.E.E.C., 2 Rue Andre Pascal, Paris 16, France.

HEILIGMAN, F., SIMON, M., and DESROSIER, N. W. 1958. Effect of irradiation on sprout inhibition, quality, and storage of potatoes. International Conference on Food Irradiation, Harwell, England, Nov. 17, 1958. Sponsored by Food and Agriculture Division, United Nations, Rome, Italy.

KONZAK, C. F. 1954. Stem rust resistance in oats induced by nuclear radiations. Brookhaven National Laboratory Bull. *1792*.

KONZAK, C. F. 1957. Genetic effects of radiation on higher plants. Quart. Rev. of Biol. *32*, 25–40.

LEOPOLD A. C. 1955. Auxins and Plant Growth. Univ. of California Press, Berkeley, California.

MIKAELSEN, K. 1954. Protective properties of cysteine, sodium hyposulfite, and sodium cyanide against radiation induced chromosome aberrations. Proc. Nat. Acad. Sci. *40*, 171–178.

NYBOM, N., LUNDQUIST, U., GUSTAFFSON, C. A. T., and EHRENBERG, L. 1953. Biological effects of X-irradiation at low temperatures. Hereditas, Lund, *39*, 446–457.

OLSON, R. W. 1958. Applications of Atomic Science in Europe and the United States in Agriculture, Food-Plant and Soils; Final Report, Project 396, European Productivity Agency, 2 Rue Andre Pascal, Paris 16, France.

Pelc, S. R., and Howard, A. 1952. Chromosome metabolism as shown by radioautographs. Experimental Cell Research, Suppl. *2*, 269–278.

Sax, K. 1957. Effects of ionizing radiation on chromosomes. Quart. Rev. Biol. *32*, 14–24.

Sereno, M. M., Highlands, M. E., Cunningham, C. E., and Getchell, J. S. 1957. Effects of irradiation and subsequent controlled storage upon composition of Maine Katahdin potatoes. Maine Agri. Expt. Sta. Bull. 563.

Skoog, F. 1935. The effect of X-irradiation on auxin and plant growth. J. Cell. Comp. Physiol. *7*, 227–270.

Smith, H. H. 1958. Radiation in the production of useful mutations. Botan. Rev. *24*, 1–24.

Sparrow, A. A. 1957. Symposium on the effects of ionizing radiations on plants. Quart. Rev. Biol. *32*, 1–56.

Sparrow, A. H., and Christensen, E. 1954. Improved storage quality of potato tubers after exposure to Co-60 gammas. Nucleonics *12*, No. 8 16–17.

Tukey, H. B. 1954. Plant Regulators in Agriculture. John Wiley and Sons, Inc., New York.

Vidal, P. 1959. Improvement of yield and quality of crops by seed irradiation. Industries atomiques *3*, 77–84.

CHAPTER 14

Radiation Effects on Animals

Human beings are the most highly evolved organisms found on the life scene and, as was noted earlier, are each an exceedingly complex organization of some 10^{30} atoms. It has been seen in the preceding chapters that increasingly complex organisms, when exposed to ionizing radiation, manifest radiation injury by increasingly complex mechanisms which form the radiation syndrome. With a highly evolved species, the measure of the results of exposure to ionizing radiation is so far expressible in terms of life and death considerations, radiation sickness—which embraces a multitude of damages to the organism, and long range effects, mainly decreased life span, physical ailments and genetical repercussions. Assuming that a man has a pool of about 10,000 genes which are equally capable of being mutated, it is calculated that the survivor of the Hiroshima blast who received 200 rads has at least one mutated gene in the chromosomes of each of his germ cells (Davidson 1957).

The exposure of the whole body of an animal to irradiation results in radiation effects found to be a function of the dose and dose-rate pattern during the exposure period. The specification of the dose-rate as a function of time is therefore a necessary item of data needed to estimate the biological effects from chronic exposure of higher animals to radiations. The external irradiation of a whole animal has three clearly defined phases of response. The early effects phase covers the period from the beginning of the exposure to approximately six months thereafter and is the time of maximum injury. In this phase the primary radiation deaths occur and non-fatal radiation sickness from mild to severe runs its course for the majority of survivors. The delayed effects phase extends from the end of the early effects phase through the life-time of the survivor. These are manifested in an increased incidence in later life of various sorts of physical ailments and reduces the life expectancy according to the severity of the dose. The long range effects extend from the first progeny of the early effects survivors to manifestations in some indeterminate future generation (Gowen 1956).

From the standpoint of biological research, estimates of animal responses to chronic whole body radiation treatments must be regarded as comparatively crude, and are at best of a gross nature.

Radiation Sensitivity vs. Organism Complexity

Man operates at the summit of a pyramid from which he views the entire life scene. To recapitulate, the most elementary forms of life which he views are the lower plant forms, those which do not reproduce by means of seeds. These include algae, fungi, mosses, and ferns. Viruses, the most minute living entities found, are extremely radiation resistant in comparison with lower plant life. Most of the plants which are familiar to man are seed bearing vascular forms. These were noted in the previous chapter to be more sensitive than lower plants to equal radiation exposures.

Animals are either of the unicellular protozoan type or multicellular metazoans. Animals may also be divided into two general groups according to the presence or absence of a backbone. The vertebrates each have a vertebral column while the invertebrates have none. The lower invertebrates include such forms as sponges, jellyfishes, and sea anemones. Radiation resistance of multicellular animal species is less than the radiation resistance of the protozoans. The helminths include the worms which infect man and his animals, inhabiting the earth's surface in soil, and in water environments. The mollusks are distinguished from other invertebrates by the usual presence of a shell and possession of a single ventral muscular foot. The arthropods comprise a large group of animals having in common the characteristics of jointed appendages, such as found in insects and crustaceans. The echinoderms are characteristic in having long or short spiny projections on their outer-body surfaces. Characteristic of these are starfishes and sea urchins. The chordates include the familiar vertebrates such as fish, frogs, reptiles, birds, and man. The lower chordates are less familiar, have no backbones, but nevertheless possess the general characteristics of the highest animal group. The lower chordates include such forms as the acorn worm, the sand worm, the sea club and related organisms, some of which appear lifeless and unresponsive in behavior, a basic chordate characteristic. The vertebrates are the higher chordates and have a spinal column composed of overlapping vertebrae, an internal skeleton of cartilage or bone or both, a greater development of head region, a dorsal nervous system, a ventral heart, eyes and nostrils on the head, together with ears in most forms, red blood with hemoglobin enclosed in corpuscles, and not more than two pairs of limbs or fins, although unpaired fins are found in some aquatic vertebrates.

The relative order of resistance or sensitivity to radiation of the living organisms seen on earth is summarized in a general way in Table 33. A typical man is destroyed by a dose of 500 rads, while viruses may survive 10,000,000 rads, other living creatures falling in between, depending on their complexity in a molecular sense.

There is a broad pattern of metabolism found in common among living entities. The metabolic pattern of complex animals includes many processes which occur in the simplest living material, for example, DNA synthesis occurs in both a virus and in man. In this chapter the effects of irradiation on those species of the animal kingdom of particular import or significance in the life of mankind, and the technological application of these radiation effects for the benefit of man will be explored.

The Loci of Radiation Damage

With the more complex animals the specialization of the cellular structure is such that the metabolic and dynamic functions involve the cooperative action of large groups of cells. Whereas in a single cell, one speaks of the microscopic location of a particular function in the structure of the cell, in the case of higher animals, there are also functions performed by multicellular organs. For example, respiration in a single cell involves the diffusion of oxygen through the cell membrane into the cell and the diffusion of carbon dioxide out of the cell, and this, in connection with certain enzyme cycles is usually sufficient to specify with some completeness the nature and details of the respiratory process. In the case of a large multicellular animal, on the other hand, one cannot consider the respiratory process exclusively in terms of the events in a single cell. In particular, large animals such as fish perform respiration by means of organs such as gills while air-breathing animals have lungs. And further, even the gills and lungs are not the only part of the organism which are directly concerned with respiration. There is also blood, which acts in part as a carrier for oxygen and for respiratory waste products.

TABLE 33

RADIATION DESTRUCTION OF LIVING ENTITIES

Organism	Generalized Killing Dose (rads)
Man	500
Higher animals	1,000
Higher plants	10,000
Lower animals	100,000
Lower plants	1,000,000
Virus	10,000,000

The general structure of large multicellular organisms may conveniently be given in terms of their organs, which are parts of the body which perform one or more particular dynamic functions. A detailed discussion of the structure and structural differences of these organisms is outside the scope of this undertaking for the following reasons. The mammals, for example, all have a brain, backbone, skeleton, lungs or gills, heart, blood vessels, and so on, all of which from species to species perform much the same function and have a great similarity in cellular structure. However, the overall appearance, overall form, and spacial arrangement of the organs within the large animal may vary considerably from species to species; and a discussion of these differences will not contribute very much to the problem at hand. Of particular importance in connection with radiation effects is the following suggestion. Although a man may have a constant weight from maturity to death, the sum total of cells within a man is constantly changing. This is directly evidenced in the growth of hair, in the replacement of skin on abrasions and cuts, and also in the fact that he is continually metabolizing a part of his own structure in order to remain alive. It is fair to say that an adult man is at best in a relatively steady state, although a man consumes ten times his body weight in food a year. Not all parts of the components of a living animal undergo the same rate of turnover. The most stable constituent of the animal is perhaps its skeleton. At the other extreme are probably the skin or epithelial and the blood cells. If we recall that one of the major effects of radiation on single-celled organisms is the inhibition of cell reproduction, it would appear reasonable that the effects on an animal due to the exposure to a particular dose of radiation may very well be related to the rate of turnover of the cells in some particular organ of the animal. Further, it is to be expected that if the sensitivity to damage by radiation with regard to cell inhibition were equal for all types of cells in the animal, then the sensitivity to radiation and the immediacy of the radiation effect should be qualitatively related to the rate of turnover of the particular cellular system involved.

ANIMAL RESPONSES TO WHOLE BODY IRRADIATION

It has been consistently found in experiments with various kinds of higher animals that as the time over which the radiation is administered is increased the total dosage necessary to produce a given severity of radiation sickness also increases. This indicates the operation of recovery mechanisms (Blair 1954).

The degree of severity of a radiation treatment that is most precisely defined and measurable is that which causes the death, within some specified period of time, of a fixed fraction of the exposed animals. The dose producing 50 per cent kill is designated as the median lethal dose, the LD_{50}. As has been noted previously, the effect of irradiation on the animal is divided into three rather distinct phases, the early, the delayed, and the long term genetical manifestations. The permanent effects of irradiation may be thought of as the delayed phase and long term effects.

Single versus Intermittent Irradiation

From results obtained by experimentation with mice, the influence of an interrupted exposure of a given dosage of radiation is as follows. A single dose of about 500 rads kills 50 per cent of the mice present. If another group of mice of the same type, age and condition are given initial doses of one half the LD_{50} and then given a second dose two, ten, and twenty days later, the results obtained are shown in the accompanying Table 34. If there were no recovery from an LD_{50}, the total radiation effects should have been the same for each group of mice (Blair 1954). The rate of recovery appears to be greatest immediately following an initial dose, then gradually diminishes as the recovery process progresses. Rate of recovery from a radiation injury has been represented in terms of the time required to achieve some given level of recovery. The choice of a particular level is somewhat arbitrary but there are a number of reasons for selecting the time to achieve 50 per cent or half recovery, or in short the "recovery half life." The recovery half lives for five species of mammals computed from experimental data by Blair are shown in Table 35. Mice are usually weaned at 30 days of age and at this stage are in their maximum growth phase. In biological terms they are producing new cells at a much more rapid rate than does the mature animal. Blair has reported that since recovery is to a large extent the process of making new cells to replace damaged ones, it is possible that the recovery half life of infant mice is appreciably shorter than that of mature mice. The point of these remarks is simply to emphasize that the range between minimum and maximum estimates of mouse recovery, for example, in Table 35 is not exclusively an expression of uncertainty but probably incorporates real differences

TABLE 34

INFLUENCE OF INTERMITTENT IRRADIATION ON LD_{50} FOR MICE[1]

Treatment	LD_{50} (rads) Total Dose Required
Single momentary dose	518
2 Doses, 2 day interval	563
2 Doses, 10 day interval	695
2 Doses, 20 day interval	736

[1] From Paterson, Gilbert, and Mathews (1947).

in response due to strains and age differences in various samplings of a species. Davidson (1957) has summarized the biological effects of whole body gamma radiation on human beings. The evidence suggests that, although there is a difference among species, a biological response scale for mouse, rat, dog, burro, and man in proper relative positions can be constructed and the recovery half life of man can be estimated with reasonable accuracy. The result from such a study indicates a recovery half life for man of between 25 and 45 days, again depending on age, condition, and type.

TABLE 35

ESTIMATED RECOVERY HALF LIFE FOR VARIOUS ANIMALS[1]

Animal	Recovery Half Life Days
Mouse	3–8
Rat	6–9
Guinea pig	7–27
Dog	14–18
Burro	20–28
Man	25–45

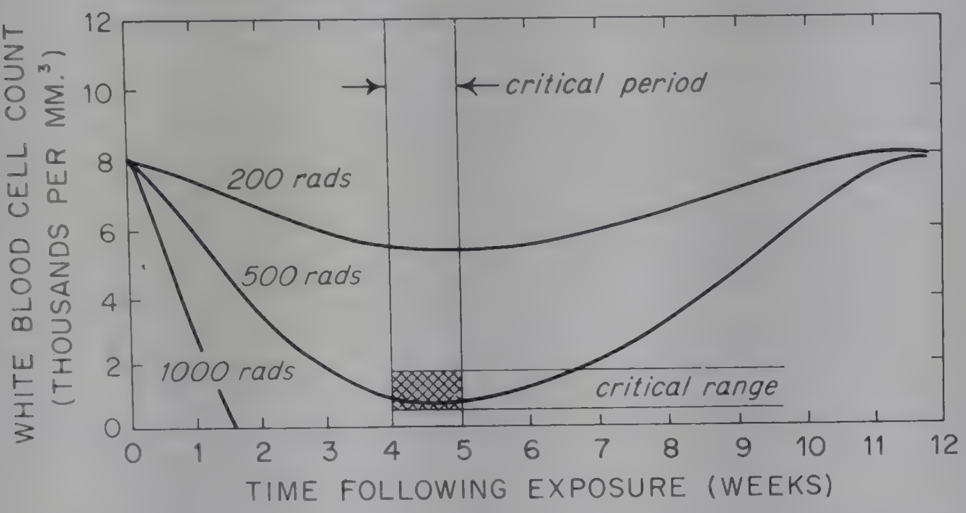
[1] Adapted from Blair (1952).

White Blood Cell Response to Radiation Sickness

The biological recovery mechanisms operating in the case of severe radiation exposure are probably similar to those that bring about the recovery from single acute doses. What is missing in the case of single dose data is a measure of recovery progress in terms of increasing tolerance to subsequent radiation. There is a general consensus of opinion that, of the various measurements possible on an irradiated animal, the circulating blood and its white blood cell content is one of the best single indices of radiation sickness. Essentially, the white blood count in warm blooded animals provides a running score on the race between the processes of deterioration and recovery in the animal organism. It appears that there is a critical range in white blood count and also a critical time that appears to be remarkably constant. This is shown in Fig. 89. The white blood count response in humans exposed to a single acute dose of radiation at three levels of severity is analyzed. These white blood cell responses were based on data from the Hiroshima bombings (Davidson 1957). Curve *A* in this figure represents an LD_{100}, death occurring within several days after exposure, to up to three weeks later. Curve *B* indicates an approximately LD_{50} dose, death occurring from the third to the sixth week, survivors exhibiting signs of recovery in about the fifth week. Curve *C* indicates dose absorptions approximately at the threshold of the lethal effect. The time for man's recovery based on white blood cell count data has been shown to be upwards of twelve weeks. The white blood cell count responses following a single LD_{50} dose in man, burro, dog, guinea pig, rat, and mouse are correlated with the recovery half life. The white blood cell count response provides a general confirmation of the pattern of species variation found as the result of dose fatality analyses.

Delayed Effects of Irradiation

The long term conditions known to be possible consequences of irradiation are not qualitatively different from similar conditions arising spontaneously in normal populations. The effect of irradiation therefore is an amplification of casual incidences of these conditions (Furth *et al.* 1957). An example is the problem of leukemia, which can be induced with a sufficient dose of radiation even in normal animals. The story of leukemia is similar to that found for other specific pathological conditions in animals, the incidence of these conditions being increased by irradiation. The pathology of delayed effects in animals is too large a subject to be adequately covered in this undertaking. In addition, the observations that known specific delayed effects of irradiation are not dissimilar from conditions arising spontaneously in a population of animals leads to a further generalization. It is considered in the aggregate that the long term somatic effects of irradiation (Table 36) are similar to the process of aging. Animal experimentation has produced unequivocal evidence of a life shortening effect at doses insufficient to induce either



Courtesy of Blair

FIG. 89. WHITE BLOOD CELLS RESPONSE IN HUMANS EXPOSED TO SINGLE ACUTE DOSES OF RADIATION RANGING FROM THRESHOLD TO LD_{100}

acute or subacute radiation syndromes. It has been found in rats and mice that at least the life shortening aspect of single yet substantial doses is different for a given dose from that found for divided doses and its magnitude may be five or more times greater. If these results are applicable to man, then it is indicated that a survivor of a single momentary dose near the LD_{50} would suffer about a 20 per cent shortening of the life expectancy of a young adult, or a loss of about ten years.

Long Range and Genetical Effects on Animals

Low level irradiation of animals may have effects on the progeny resulting from sexual reproduction. Because of the great complexity of the animal organisms, particularly the mammals, the effect of radiation is generally destructive. The overall pattern of effects is summarized in Table 36. Starting with a fertilized egg, 50 successive cell divisions by mitosis yields all the cells which when fully grown yield the adult organism. Radiation damage to an adult organism capable of successful reproduction finds various ramifications. Pertinent in the long term effects would be the damage inflicted during the process of meiosis, during which the production of sperm and egg occur in the male and female. At this critical stage, long term genetical effects may be anticipated in distant progeny. Other irradiation syndromes are manifested in tumors, radiation burns, and radiation sickness in general. While no useful application of whole body irradiation has been found due to its extremely damaging effects on the highly complex mammal organization, selective irradiation of reproductive organs in animals is employed in the search for useful mutations. The use of point irradiation in cancer therapy is also noteworthy.

TABLE 36

SUMMARY OF RADIATION EFFECTS ON ANIMALS

Somatic Effects:	*Genetical Effects:*
Radiation burns	Lethal mutants
Radiation sickness	Monstrosities which live
Leukemia	Useful mutants
Tumors	
Physical ailments	
Malformities	
Shortened life span	

That mutations can be induced by irradiation is well established. Classic work with irradiated Drosophila has revealed a great variety of genetic effects. It must not be supposed that all are necessarily useless. The probability of a useful occurrence is more remote with complex animals. This does not preclude the possibility that further studies will produce useful applications, however.

Radiation Protective Additives

There are encouraging results from laboratory experiments with animals which anticipate the eventual development of substances that would in effect permit survival of an animal to what otherwise would be a lethal dose. Studies on mice have revealed that the introduction of small amounts of the amino acid cysteine produces a marked increase in the number of survivors of an otherwise lethal dose. The mechanism appears to lie in a favorable modification of the recovery process rather than the establishment of a partial immunity condition. Spectacular results have been found by introducing unirradiated bone marrow into vertebrates which have received lethal doses. These techniques are extremely important findings and their future application encouraging. However, the magnitude of the effect that may be anticipated for radiation protective agents or additives does not suggest that their development would drastically alter the character of radiation syndromes or entirely prevent radiation damage.

Summary of Biological Effects on Animals

The early response of mammals to chronic or intermittent gamma radiation can be represented as a reparable effect, the recovery being proportional to the magnitude of the effect. Recovery of tolerance to additional irradiation does not correspond chronologically to the recovery of fitness. Performance of life responsibilities by an animal is altered. Following irradiation, vulnerability to infectious diseases is at a maximum. Various mammals exhibit significant differences between species in recovery following sublethal radiation doses. Analysis of data on the white blood cell responses due to irradiation also shows a pattern of species difference.

For short term (less than a week) exposures the cumulative dose can be used to estimate the radiation effects. With longer exposures of equal accumulated doses the recovery processes play an important part in determining the overall results of multiple exposures. The consequence to human beings will be discussed in the section dealing with health physics and personnel protection.

It is seen that radiation directly influences the longevity or life span which an animal will experience, and this is proportional to the dose absorbed.

The effects of irradiation on progeny of mammals appears as a delayed effect and will not be completely understood for many generations of mankind yet to come.

The striking feature of exposures of animals to ionizing radiation is the apparent inability of the animal to sense the exposure. In the evolution of humans and mammals, the demand for a sense to detect ionizing radiation appeared not to be an evolutionary requirement. The ability of some organisms to sense low energy (visible) radiation, however, is self evident.

Some fascinating information has been uncovered relative to the sensitivity of various humans to radiation damage. It has been found that diabetics are as a group less sensitive to radiation damage than "normal" counterparts. Glucose has the ability to inactivate hydroxyl radicals produced by the irradiation of aqueous solutions as noted earlier.

The effects of highly intense radiations intentionally inflicted or suffered through error are being carefully studied in many laboratories around the world. The best guide for protection from such effects is the experience gained from damage done to man and animals in the past; in the future the deleterious consequences from overexposure to radiation should be encountered only in those unavoidable accidents or in purposeful radiotherapy. Many of the manifestations of acute radiation effects are attributable to the injury of the self-replenishing tissues in the animals. The haematopoietic system appears especially important in this regard. Delayed effects range from the shortening of the life expectancy or span to the development of specific changes such as cancers. Tumor induction can occur with the deposition of radioactive elements in tissue, and may result from the direct injury by radiation or indirectly through hormonal or other unknown mechanisms. Bone tumors are associated with absorption of radioactive substances that have a metabolic similarity to calcium such as strontium-90. Chromosomal injury in developing germ cells gives rise to genetical alterations in the succeeding generations as noted above. Radiation has effects on the reproductive system and on the foetus. Irradiation during the life of the foetus in the mother may give rise to a high frequency of developmental abnormalities. Spermatogenesis is disturbed in animals given radiation exposures approximately 10 times that industrially permitted over a period of three years (Davidson 1957). In discussing radiation damage or radiation effects on the genetical mechanisms, it is useful to distinguish between the somatic and the genetic aspects. Somatic damage does not affect the germ cells and hence is not transmitted from one generation of animal to the next. No matter how extensive the damage at any one time the next generation is spared the effects. Some types of genetical damage follow precisely the pattern of activity seen in somatic damage. Dominant lethal and dominant sterility factors are completely eliminated from populations in the generation following the exposure. The definition of reasonable levels of tolerance to exposures of ionizing radiation is an uncertain matter. It is thought that the exposure of large numbers of populations to ionizing radiation will not evidence itself in monstrosities in future generations. The natural selection which occurs widespread in nature acts to eliminate deleterious mutants which would be monstrosities. On the general life scene, the supply of genetical material passed from one generation to another gives rise to remarkably normal individuals considering the multitude of opportunities for deleterious mutations to occur.

While the irradiation of the whole body of mammals leads to radiation syndromes and the radiation tolerance of the mammals appear low, at best in the hundreds of rads, the fact that it is possible to employ many times this quantity of radiation to isolated areas of the mammal organism without serious damage has been widely applied. Perhaps the most significant of these applications is that dealing with the destruction of cancerous growths in mammals using pinpoint irradiation techniques. Tissues are destroyed locally and with due caution do not inflict injury to other cells of the body. Cancerous growths in humans, for example, can be destroyed by the judicious application of between 6,000 and 9,000 rads of radiation.

While it would appear that the irradiation of whole animals offers no benefit, this is not the case. It is a fact that mammals are infested with a large number of parasites which can be destroyed with radiation. For example, of all the human autopsies made and recorded in the United States, upwards of 15 per cent have been found to be infected with worms (Belding 1956) and these gained entry through the vehicle of food and drink. The widespread infection in mankind in general can be seen from Table 37, which considers only selected helminths acquired by man. The incidence and geographical distribution of such infestations is alarming. While equal data on other parasites is obtainable, it suffices to note that their hold on mankind is substantial. Mankind may control such infestations of himself through control of his animals, his plants, and his water supply. The application of radiation to the destruction of insects and parasites which plague man's food supply appears most challenging and rewarding (Proctor *et al.* 1954).

Radiation Effects on Animal Parasites

The control of many parasitic infections is within the bounds of present radiation technology and offers tremendous possibilities for alleviating the widespread and insidious public health problems of the present time. Certainly the order of magnitude of the problem is staggering, TABLE

SOME IMPORTANT HELMINTHIC DISEASES

Causative organism	Form in which ingested by man	Disease produced in man
Trichinella spiralis	Cysts in raw or undercooked pork.	Trichinosis
Clonorchis sinensis	Metacercariae in fresh-water fish.	Clonorchiasis (Chinese liver fluke).
Ascaris Lumbricoides	Ova in raw vegetables (principal mode of transmission is is infected soil).	Ascariasis. (Intestinal obstruction in children, pneumonia).
Fasciolopsis buski	Metacercariae on raw fruit or edible water plants.	Fasciolopsiasis (intestinal fluke).
Taenia saginata	Cysticerci in meat	Beef tapeworm
Taenia solium	Cysticerci in pork	Pork tape worm; cysticercosis in tissues.
Diphyllobothrium latum	Plerocercoid larva in fresh-water fish.	Fish tapeworm anemia resembling pernicious anemia.
Paragonimus westermani	Crabs or crayfish containing ing metacercariae.	Paragonimiasis (oriental lung fluke).
Fasciola hepatica	Encysted cercaria in water-cress, lettuce.	Fascioliasis (sheep liver fluke).

---

* These numbers have reference to Belding, D. L.: *Textbook of Clinical Parasitology*, Second Ed., New York:

** Stoli, N. R.: "This Wormy World," *The Journal of Parasitology*, 1947, Vol. 33, p. 15.

yet constructive steps to remedy the situation can be taken (Gomberg and Gould 1956). As will be shown, the radiation resistance of parasites as a group is much less than that of bacterial spores, and of the order of sensitivity of nonresistant vegetative cells.

To consider helminths first, their infestations are of significance in public health and of economic import as well. The worms can be divided into three groups, the roundworms (nematodes), the tapeworms (cestodes), and the flukes (trematodes) (Belding 1956).

Ascaris, the common roundworm, is infective through its ova, and matures in man's intestinal tract. *Trichinella* is taken into the body in the larval form, mainly from pork, develops to maturity in the small intestine, and spews young larvae internally, which eventually encyst themselves in muscle. There is no known remedy for this infection, once encysted in

37

OF MAN ACQUIRED THROUGH FOOD

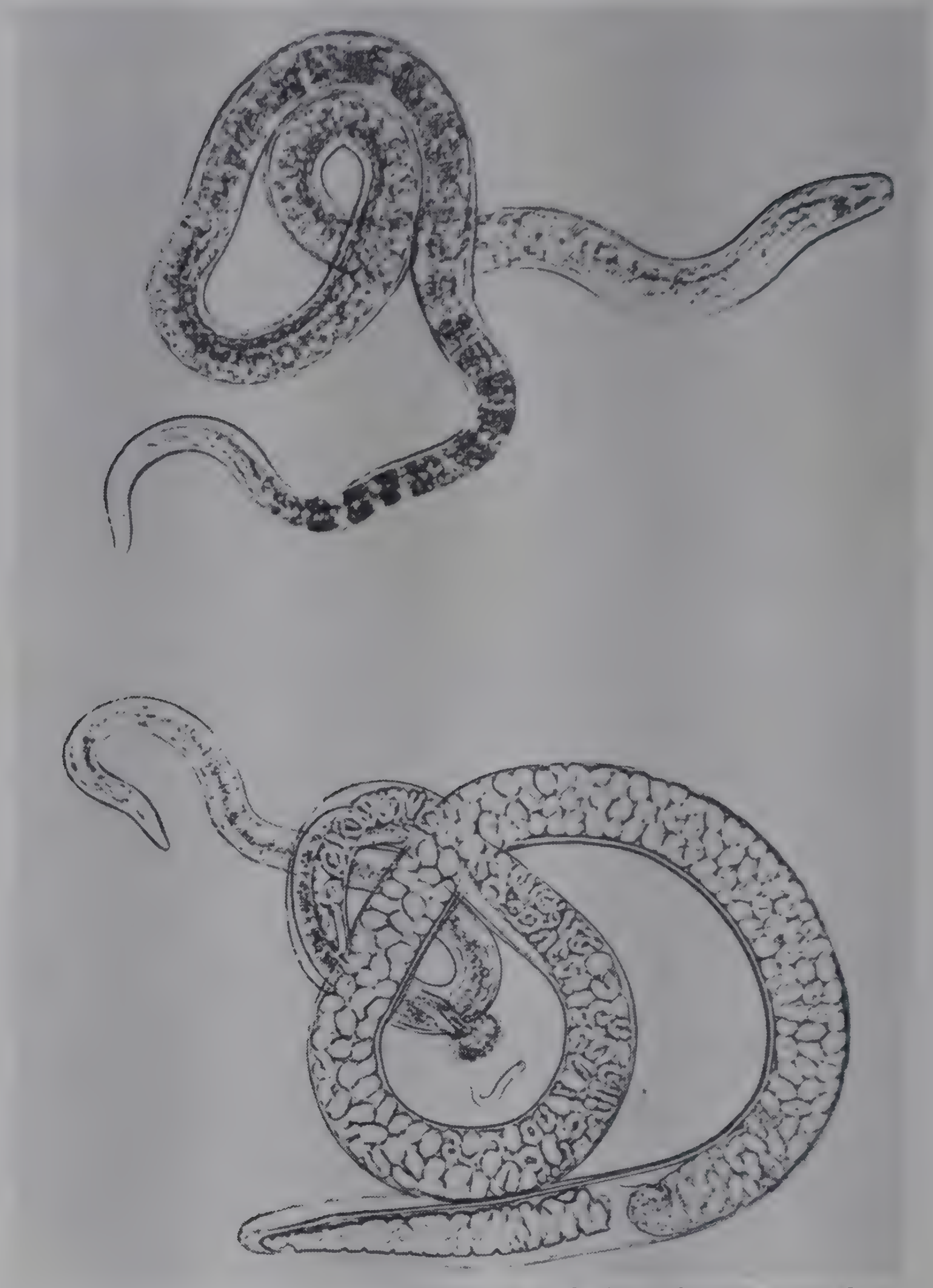
Natural host	Incidence and geographic distribution
Hog (rat, man, carnivorous animals).	Cosmopolitan. In 16 per cent human autopsies in U. S. (p. 370).* In U. S. 1.5 per cent of all hogs are infected (p. 370).* 27.8 million people.**
Man, dog, cat, hog (snails and fish are intermediate hosts).	Japan, China, Formosa, Korea, French Indo-China. "Disease is equally prevalent in the poor and the prosperous, in the illiterate as well as the educated," because of custom of eating raw fish (p. 650).* 19 million people.**
Hog, man	Cosmopolitan. In surveys of U. S. Southern States, 1 to 60 per cent in children (p. 447).* In Shantung, China, 23 to 44 per cent in children. 644.4 million people.**
Man, hog (snail is intermediate host).	Central and South China. 10 million people.**
Man (cattle are intermediate hosts).	Cosmopolitan. 0.38 per cent of persons infected in Peiping, China. Incidence in cattle: U. S. 0.37 per cent, Holland 3.3 to 5.6 per cent, S. Africa 5 per cent (p. 578).* 38.9 million people.**
Man (hog is intermediate host).	Cosmopolitan. Incidence of tapeworm in man: Russia 0.2 to 1.5 per cent; incidence of cysticercosis in man: 2.9 per cent Mexico (p. 575).* Incidence in hogs: South Africa, 25 per cent; Mexico, 4.3 per cent. 2.5 million people.**
Man, dog, cat (fish is intermediate host).	North America, Europe, Tropical Africa, North Asia, South Asia. In Finnish Army 14.5 per cent (p. 548).* About 20 per cent of pickerel and pike in Lake Winnipeg and Lake Manitoba are infected (p. 549).* 10.4 million people.**
Man, dog, cat, many other animals (snail and crab or crayfish are intermediate hosts).	Japan, Korea, Manchuria, Formosa, China, other countries of Orient; Africa, South America. Adult parasites have been found in mammals in Canada and many states of U. S. 3.2 million people.**
Man, sheep (snails are intermediate hosts).	Cosmopolitan, in sheep-raising areas of world. Incidence in Estonia 57 per cent in sheep, 72 per cent in cattle; in Transcaucasia, 48 per cent in sheep; in Southern Texas, 21 to 32 per cent of rabbits (reservoir hosts for new infections) (p. 629).* 100,000 people.**

Appleton-Century-Croft.

human tissue. The pork tapeworm, *Taenia solium,* is found in its encysted larval stage in pork commonly referred to as "mealy." The fish tapeworm, *Diphyllobothrium latum,* also is found as an encysted larvae in the flesh of fish, developing tapeworms in the intestinal tract of man. The latter parasite is apparently becoming entrenched in North Central America.

The liver fluke, *Fasciola hepatica,* is transmitted through plants and animals which man tends to eat raw or insufficiently cooked, permitting the larvae to be ingested in an infective stage.

Destruction of Worms.—Radiation damage to helminths can be considered in terms of cell life cycle stages. The eggs of roundworms have been found to be radiation sensitive (Wharton 1957). Exposures to 50,000 rads leaves few if any survivors, and these survivors usually have



Courtesy of U. S. Army Quartermaster Corps

FIG. 90. RADIATION EFFECTS ON *Trichinella spiralis*

Destruction of the organisms seen when exposed to 50,000 rads. Normal organism at bottom of photograph

lost their infectivity. The irradiation of adult worms produces females that are sterile after exposures to 25,000 rads and distorted morphologically with 50,000 rads. *Heterodera,* a nematode infecting root crops, has been found to yield sterile eggs when adults are irradiated at 20,000 rads. With *Trichinella,* 15,000 rads sterilizes the females in the adult stage, and 25,000 rads destroys the larval ability to infect animals. Control of infectivity would seem possible at 50,000 rads. The changes which occur in *Trichinella* following irradiation can be seen in Fig. 90, which demonstrates the morphological manifestations of ionizing radiations internally in the worm.

The striking feature of gamma radiation is its ability to penetrate whole tissues, being capable of penetrating large whole animal carcass. Considering the dose requirements to eliminate infectivity, such treatments are economically within reach. The biological and technical control of most worms transmitted in food is feasible.

Destruction of Protozoa.—The irradiation of protozoa has marked and destructive effects on the organisms. Many of the same factors which influence the radiation effects in bacteria are operative with protozoa, although there are variations.

It is well established that irradiation can cause death to protozoa. Low doses may immobilize, prevent mating, reduce reproductive rates, and change morphological characters (Wharton 1957).

TABLE 38

RADIATION EFFECTS ON PARASITIC PROTOZOA[1]

Organism	Dose (rads)	Effect
Trypanosoma brucei	10,000	Reduced infective power
T. gambiense	12,000	Failure to infect
	100,000	Killed
T. cruzi	15,000	Reduced infective power
Plasmodium gallinaceum, endothelial stages	700	No effect on infectivity
“ “ sporozoites	8,000	Failure to infect
“ “ trophozoites	20,000	Failure to infect
P. malariae trophozoites	5,000	Failure to infect
	20,000	Killed
P. lophurae trophozoites	16,000	Failure to infect
Eimeria tenella oöcysts	14,000	Failure to infect
E. tenella	15,000	Reduced infective power and distinct decrease in severity of infection
Adelina deronis	1,000	Abnormal growth
Endamoeba histolytica	150,000	Abnormal growth
E. histolytica trophozoites	60,000	Partial inhibition of growth
	120,000	Infectivity unchanged
	600,000	Killed

[1] Adapted from Kimball (1954).

The loss of infective capacity of parasitic protozoa is a useful phenomenon, and is brought about by dosages much lower than those required to kill the organisms. The infectivity of *Trypanosoma cruzi* was found by Emmett (1950) to be decreased by 10,000 rads and destroyed by 100,000 rads, yet neither the apparent vitality nor the reproductive power of the organisms was affected. At doses of 150,000 rads, Sadun found that *Endamoeba histolytica* trophozoites were not reduced in infectivity but did have stunted growth patterns. The effects of irradiation are dependent upon the life-cycle of the organisms. *Plasmodium malariae* given 8,000 rads at the sporozoite stage are killed, whereas trophozoites require 20,000 rads for elimination, as reported by Bennison and Coatney (1945). Brown has shown that *Euplotes* cysts are killed by 400,000 rads while the trophozoites require nearly 500,000 rads.

The lethal effects of ionizing radiation on protozoa may take several paths. The dose applied may be sufficient to kill the cells outright, or yield death after considerable time without cell division occurring. Death may occur shortly after division, or after several divisions, or following sexual processes.

Cytolysis of *Pandorina morum* occurs with approximately 400,000 rads, and to *Paramecium* by 500,000. The amoeba *Polmyxa* requires more than 100,000 rads to kill, although morphological alterations occur at this level.

Characteristic responses of parasitic protozoa to ionizing radiations reported in the literature have been tabulated by Kimball, a portion of which is shown in Table 38.

RADIATION EFFECTS ON INSECTS

In comparison with warm blooded animals, insects are radiation resistant. In comparison with bacteria, insects at the most have about the resistance of vegetative cells. There is a wide range in the sensitivity of different orders and species of insects to radiation, and the stage of development in the life cycle is equally important. A major problem is posed in establishing the lethal dose of radiation in different species, within species, and even ages within the stage of a species. Nevertheless, it is possible to arrive at some useful concepts relative to the effects of radiation on insects.

Radiation often fails to cause immediate death. If a series of eggs, larvae, pupae, and adults of a species were introduced into a gamma field, recovered, and observed, it would be seen, if the dose rate were not overwhelmingly strong, that the insect fails to pass properly through its usual stages. Death may occur during the process of moulting, for example. Long term effects appear as lethal or developmental defects occurring some time after the test organism was exposed. Irradiated eggs may yield non-viable larvae. Long term effects may also be manifested in deformed organs and tissues if the insect survives.

Hilchey (1958) noted that the irradiation interfered with reproductive capacities in at least three ways. One is the interference of developmental processes involved in the growth of reproductive organs. *Aedes aegyptia* larvae or pupae exposed to radiation may fail to develop ovaries. Another effect of irradiation is a direct effect on gametes in the body of a normal adult. And third, the adult may fail to take part in normal sexual activities, perhaps to be recognized as a behavioral breakdown. The doses required to inflict reproductive failures are available between orders and species of insects.

The length of life of an insect is usually reduced with increasing dose administered. However, there are exceptions. It has been suggested by Sullivan and Grosch (1953) that the treatment may injure the metabolic efficiency of test species, and this damage slows the life processes in the insect which results in a longer life span. It would appear that this may occur for example with *Tribolium*, *Drosophila*, and *Hadrobracon*.

The rate of respiration of insects is also related to the dose administered. While low doses have been noted to increase slightly the oxygen consumption, generally the higher the dose the lower the rate. Food intake is usually reduced following irradiation, as is the frequency of feeding.

In overall pattern, a given dose of radiation appears to have a greater effect on anabolic processes than on the catabolic.

Radiation can interfere directly with mitosis in cells, and as with whole insects, the stage of development influences the responses. Cells in the process of differentiation show a higher resistance to radiation damage than those cells in mitosis. That radiation can cause mutations in insects is well established, and is a common tool in studies with *Drosophila*, for example, where important information on populations is being obtained. The environment in which insects are irradiated interacts with the effects of the irradiation. High oxygen tension increases radiosensitivity of insect cells; the temperature at which irradiation takes place is also significant.

Destruction of Insects

The first known application of ionizing radiation in the disinfestation of a commodity was found in the cigar industry, where the attack of the cigarette beetle caused great economic loss (Hilchey 1957). This work was done in 1912 and it was shown clearly that eggs, larvae, and adults were killed by irradiation. Because of the poor radiation sources available at that time, the process was abandoned; with the present sources such applications are now promising. The destructive effects of insects on dry foods are enormous, and, while chemical disinfestations are succesful in some instances, radiation should enjoy a favored use in the treatment of packaged products.

While it is indicated in Table 39 that the radiation resistance of insects is variable, and that the stage of development of the insect in part controls its radiation sensitivity, such factors are not within the control of a processor and doses must be administered with the possibility that there will be a range of resistances in the insect population, and that a dose

TABLE 39

RADIATION EFFECTS ON SELECTED INSECTS

Organism	Common name	Dose (rads)	Effects
Acanthoscelides obtectus	Bean weevil	1,000	Eggs killed
		1,500	Larvae killed
		3,000	Pupae killed
		20,000	Adults killed
Sitophilus granarius	Granary weevil	10,000	Killed eggs, larvae
		20,000	Killed adults
Sitophilus oryza	Rice weevil	20,000	Kills all forms
Tribolium confusum	Confused flour beetle	50,000	Eggs, larvae, pupae killed
		200,000	Adults killed
Oryzaephilus surinamensis	Saw toothed grain beetle	50,000	90 per cent eggs killed
		100,000	Larvae killed
		125,000	Adults killed
Lasioderma serricorne	Cigarette beetle	50,000	Eggs killed
		100,000	Pupae killed
		100,000	Adults killed
Rhizopertha dominica	Lesser grain borer	25,000	Eggs killed
		50,000	Pupae killed
		75,000	Adults killed
Tenebrio molitor	Yellow meal worm	70,000	Pupae killed
		200,000	Kills all forms
Drosophila melanogaster	Fruit fly	1,000	Eggs killed
		5,000	Larvae killed
		12,000	Pupae killed
		200,000	Adults killed

adequate to destroy the most resistant forms is required. Estimates of this dose requirement range from 100,000 to 150,000 rads. With the progress being made in source design and radiation economics, it would be reasonable to conclude that the cost of the application of 200,000 rads to a package of food may soon be in the vicinity of a tenth of a cent a pound.

It has been found by Hilchey (1958), using radioactive tracers to follow the feeding patterns of irradiated insects, that there is some feeding by the insect even though it will die in a few days or a week. And, this insect, though crippled, is still able to cause some economic damage after exposure to doses near 25,000 rads. The application of higher levels of irradiation reduces this problem to insignificance.

Insect Penetration of Packaging Materials

Insects attempt to penetrate packages regardless of whether the package contains foodstuff or whether the foodstuff is suitable for the insects development. An analysis by Tripp (1957) lists the following reasons for insect invasion of packages:

—in search of food (example—flour beetle)
—to form pupal cells for transformation from larva to adult (example—the cadelle)
—to lay eggs (example—granary weevil)
—to eat the packaging material itself (example—termites)
—because some food is present on outside of package
—for unknown reasons.

The ability of insects to penetrate packages varies with the species, some being capable of penetrating 0.002 inches of aluminum foil which has been creased (Proctor *et al.* 1954). Once such a penetration has been made, entrance by many other insects and organisms may occur. The cadelle is noteworthy in this regard and, being widely distributed, usually can be anticipated where foodstuffs are commonly stored.

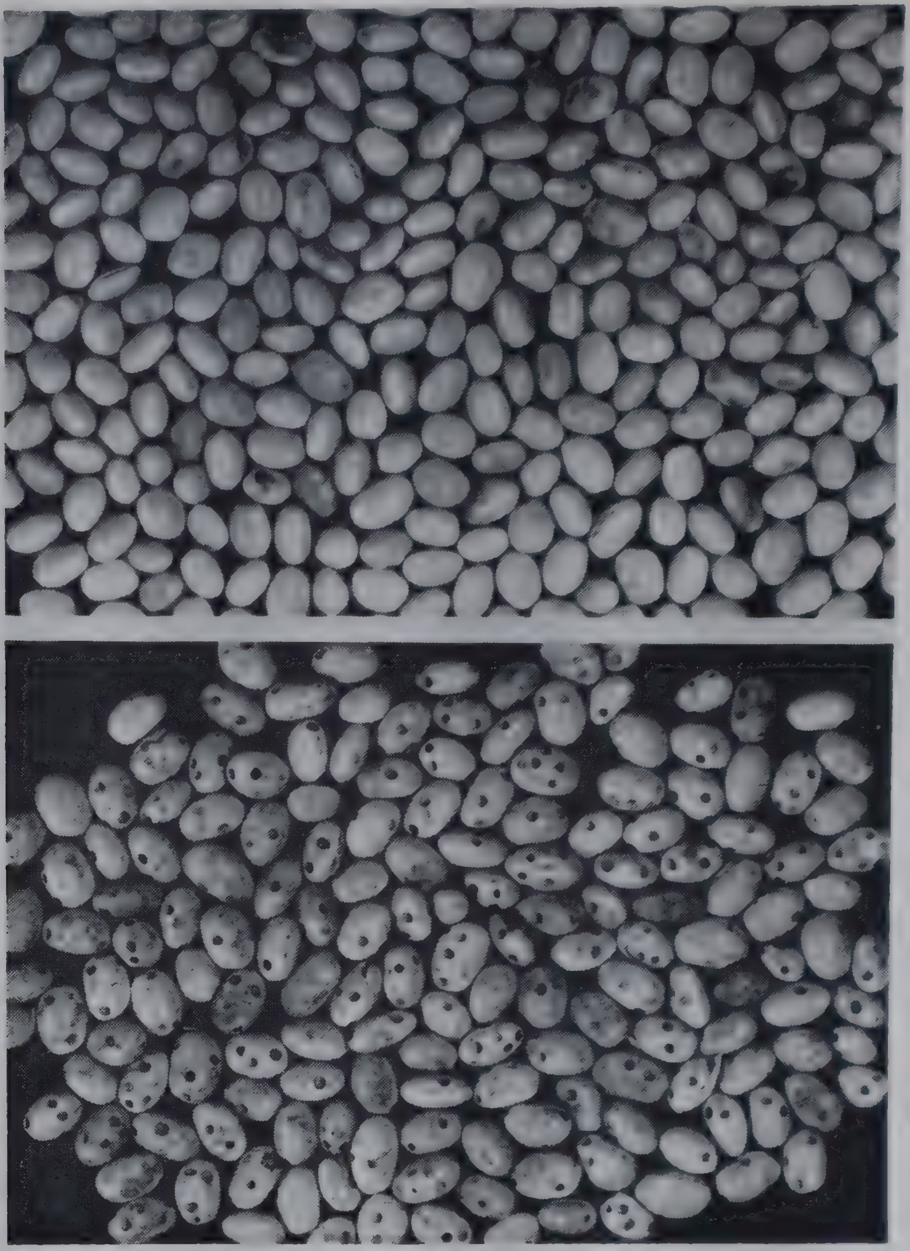
Those insects incapable of penetrating a package are able to deposit eggs in and along folds and creases which are an inherent part of a package. When small larvae develop, they may then penetrate packages through perforations too small to be seen by the unaided eye.

Glass and tin plated steel containers are resistant to insect penetration, as are certain wax coated barriers, if undamaged. Most other packaging materials may be penetrated by insects, under certain conditions.

The incorporation of insecticides and insect repellents on and in films reduces insect penetration into packages. One such treatment found effective is a mixture of pyrethrins and 40 mg. piperonyl butoxide per square foot of surface. While irradiation of such packages with 500,000 rads does not alter the insect resistant properties of pyrethrum, doses over four million rads reduce its useful properties.

Infestation of grains and fruits starts in the field and is carried to the storage or processing plant. There are a number of methods of destroying insects in stored food products. These include chemical treatments, high frequency heating, and sonic treatments. Chemical fumigation is the method currently in widespread use. Yet, for fumigants to be effective, the toxic substance must be present for substantial periods of time, and in sufficient concentrations. While this method is suitable for large storage bins or rooms, the use of fumigants for packaged foods has not been satisfactory. Fumigants are usually incapable of penetrating sealed packages, and if introduced into the food within the package, the toxic additive might not be dissipated before the product could be consumed. Radiation has special advantages in these respects.

The dose of radiation administered to control insects is dependent in part on the effects on the food commodity itself and its radiation sensitivity. With wheat, upwards to 400,000 rads can be given without serious



Courtesy of U. S. Army Quartermaster Corps

FIG. 91. INSECT DISINFESTATION OF BEANS WITH LOW LEVEL GAMMA IRRADIATION

Treated sample at top of photograph

damage to flavor; bread made from the flour produces a satisfactory loaf. The irradiation of flour yields some off flavors in the 50,000 rad level, although bread and cakes produced from the flour are still satisfactory. With higher doses and current technology, increasingly undesirable characteristics develop. Bread and cakes tend to become gelatinous and brown in appearance. Oats irradiated with up to a million rads are relatively unaltered or damaged in stability or functional use. Spices irradiated to 100,000 rads show but slight effects. Cinnamon does develop a slight bitterness at this level. Yet with present knowledge insects can be destroyed by irradiating grains, flour, spices, and other dried foods without impairing their usefulness (Fig. 91).

The use of radioactive tracers to study the metabolism of plants and animals has yielded information leading to very practical results. The observations that large amounts of radioactive phosphorus fed to the common house fly did not seem to injure the insect but caused sterility led to the rather spectacular experiments in which male screw worm flies were also sterilized, using gamma radiation. The female mates once a season. The encounter of a radiation sterilized male with a fertile female yields sterile offspring. The screw worm fly has inflicted an economic loss in the southern United States estimated to be about 15 million dollars annually. Means of eradication were less than satisfactory (Baumhover *et al.* 1955).

The release of millions of male flies sterilized by radiation was attempted in 1955 on an isolated island in the Caribbean area and led to a drastic reduction in the usually anticipated population of flies the next season. After four years of controlled release of male steriles, the screw worm fly has been reduced to an insignificant population. With this success, large scale efforts are underway in the southern United States to attempt to exploit the findings on this continent. It appears that the opportunities of usefully employing radiation on animals are limited only by man's imagination.

BIBLIOGRAPHY

ALICATA, J. E. 1951. Effect of roentgen radiation of *Trichinella spiralis*. J. Parasitology 37, 491–495.

ASHBY, W. 1948. The span of life of the red blood cells. Blood 3, 486–500.

BACHOFER, C. S., and PAHL, G. 1955. Influence of extended temperature treatments on recovery of X-irradiated Ascaris eggs. Radiation Research 2, 50–56.

BAUMHOVER, A. H., GRAHAM, A. J., BITTER, B. A., HOPKINS, D. E., NEW, W. D., DUDLEY, F. H., and BUSHLAND, R. C. 1955. Screw worm control through release of sterilized flies. J. Econom. Entomology *48*, 462–466.

BEAM, C. A. 1959. The influence of metabolism on some radiation effects. Radiation Research Suppl. No. 1, 372–390.

BENNISON, B. E., and COATNEY, G. R. 1945. Inactivation of malarial parasites with X-rays. U. S. Public Health Service. Health Rept., No. 60.

BELDING, D. L. 1956. Textbook of Clinical Parisitology. Second Ed., Apple-ton-Century-Croft Publ. Co., New York.

BLAIR, H. A. 1954. Biological Effects of External Radiation. McGraw-Hill Book Publ. Co., New York.

BLAIR, H. A. 1952. A formulation of the injury, life span, dose relations for ionizing radiations. Rept. Atomic Energy Project UR-206. University of Rochester, Rochester, N. Y.

BLAIR, H. A. 1954. Recovery from radiation injury in mice and its effect on LD_{50} for durations of exposure up to several weeks. Atomic Energy Project Report UR-312. Rochester, New York.

Brown, M. D., Luck, J. M., Cheets, G., and Taylor, C. V. 1933. The action of X-rays on *Euphates Taylori* and associated bacteria. J. Gen. Physiol. *16*, 397–403.

Brownell, L. E., Nehemias, J. V., and Bulmer, J. J. 1955. The design of a gamma irradiation facility for the control of insect infestation in flour, meal, or grain. U. S. Atomic Energy Commission Doc. No. AECU-3050.

Brues, A. M. 1956. Modes of radiation injury; bone and carcinogenesis; and sterility and foetal damage. Progress in Nuclear Energy Series VI. Biological Sciences p. 1–13.

Butler, J. A. V. 1959. Changes induced in nucleic acids by ionizing radiations and chemicals. Radiation Research Suppl. No. 1, 403–416.

Carling, E. R. 1956. Modes of radiation injury: medical aspects. Geneva Conference on Peaceful Uses of Atomic Energy, Paper No. P/450.

Carter, T. C. 1956. Genetics—fundamental effects of radiations and the hazards to man. Progress in Nuclear Energy Series VI. Biological Sciences, p. 13–47.

Comfort, A. 1959. Natural aging and the effects of radiation. Radiation Research Suppl. No. 1. 216–234.

Conard, R. A., Meyer, L. M., Robertson, J. S., Sutow, W. W., Wolins, W., and Hechter, H. 1959. Effects of fallout radiation on a human population. Radiation Research Suppl. No. 1, 280–295.

Davidson, H. O. 1957. Biological Effects of Whole Body Gamma Radiation on Human Beings. The Johns Hopkins University Press, Baltimore, Md.

Doan, C. A. 1954. The white blood cells in health and disease. Bull. N. Y. Acad. Med. *30*, 415–428.

Emmett, J. 1950. Effect of X-radiation on *Trypanosome ornzi.* J. Parasitol. *36*, 45–47.

Furth, J., Upton, A. C., and Kimball, A. W. 1959. Late pathological effects of atomic detonation and their pathogenesis. Radiation Research Suppl. No. 1, 243–264.

Gaulden, M. E., and Nix, M. 1950. Effects of oxygen tension on X-ray induced mitotic inhibition. Genetics *35*, 665–669.

Gomberg, H. J., and Gould, S. E. 1953. Effect of irradiation with cobalt-60 on trichina larvae. Science *118*, 75–78.

Gomberg, H. J., and Gould, S. E. 1956. Prevention of human helminthic diseases by gamma radiation of food. Proc. Intern. Conf. Peaceful Uses of Atomic Energy, *15*, 251–256.

Gowen, J. W. 1956. Effects of whole body exposure to nuclear or X-ray energy on life span and life efficiency. Geneva Conference on Peaceful Uses of Atomic Energy, Paper No. P/256.

Gray, L. H. 1954. Some characteristics of biological damage induced by ionizing radiations. Radiation Research *1*, 189–196.

Grosch, J. W., and Stadler, J. 1952. Fate of radiostrontium fed to Hadrobracon females. Science *123*, 141–143.

Harrington, N. J., and Koza, R. W. 1951. The effect of X-radiation on desoxyribonucleic acid and on the size of grasshopper embryonic nuclei. Biol. Bull. *101*, 138–144.

Hassett, C. C., and Jenkins, D. W. 1952. Fission products for insect control. Report 149, Chemical Corps. Med. Lab., Army Chemical Center, Maryland.

HILCHEY, J. 1957. Action of ionizing radiations on insects. Chap. 28. Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

HOLLAENDER, A. 1954. Radiation Biology. McGraw-Hill Book Publ. Co., New York.

KIMBALL, R. F. 1954. The effect of radiation on protozoa and the eggs of invertebrates other than insects. Chap. 8, Vol. 2, Radiation Biology, Editor, Hollaender, A. McGraw-Hill Book Co., New York.

KOHN, H. I. 1959. The late effects of ionizing radiation. Radiation Research Suppl. No. 1, 235–242.

LATARJET, R., EPHRUSSI-TAYLOR, H., and REBEYROTTE, N. 1959. On the target size of a transforming factor based on X-ray inactivation. Radiation Research Suppl. No. 1, 417–430.

LORENZ, E., and CONGDON, C. C. 1956. Modification of acute and chronic irradiation injury in experimental animals by injection of bone marrow and tissues originating from hematopoietic organs. Proc. of Fourth International Conference of the International Society of Hematology.

NICKSON, J. J. 1952. Symposium on Radiobiology—The Basic Aspects of Radiation Effects on Living Systems. John Wiley & Sons, Inc., New York.

PATERSON, E., GILBERT, C. W., and MATHEWS, J. 1947. Time-intensity factors and whole body irradiation. Brit. J. Radiology 25, 427–433.

PROCTOR, B. E., LOCKHART, E. E., GOLDBLITH, S. A., GRUNDY, A. V., TRIPP, G.E., KAREL, M., and BROGLE, R. C. 1954. The use of ionizing radiations in the eradication of insects in packaged military rations. Food Technol. 8, 536–544.

PROSSER, C. L., PAINTER, H. I., BRUES, A. M., JACOBSON, L. O., and SWIFT, M. N. 1947. Clinical sequence of physiological effects of ionizing radiation in animals. U. S. Atomic Energy Commission Rept. 611, Oak Ridge, Tenn.

RUSSELL, L. B., and RUSSELL, W. L. 1956. Hazards to the embryo and foetus from ionizing radiations. Geneva Conference on Peaceful Uses of Atomic Energy, Paper No. P/77.

SACHER, G. A. 1955. A comparative analysis of radiation lethality in mammals exposed at constant average intensity for the duration of life. J. National Cancer Inst. *15*, 1125–1144.

SADUN, E. H., ANE, J. N., FULLER, F. W., and LEWIS, R. 1950. The effect of X-rays upon the growth and infectivity of *Endamoeba histolytica*. Am. J. Tropical Med. *30*, 635–637.

STOLL, N. R. 1947. This wormy world. J. Parasitology, 33, 15–21.

SULLIVAN, R. L., and GROSCH, D. C. 1953. The radiation tolerance of the adult wasp. Nucleonics *11*, No. 3, 21–22.

TAHMISIAN, T. N. 1956. Studies on the biological basis of radiosensitivity. Proc. Intern. Conf. Peaceful Uses of Atomic Energy *11*, 256–263.

TRIGG, G. E. 1957. Packaging of irradiated foods. Chap. 28. Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

WHARTON, D. R. A. 1957. Action of ionizing radiations on protozoa. Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

WHARTON, D. R. A. 1957. Action of ionizing radiations on helminths. Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

WHITING, A. R. 1954. The effects of oxygen on the frequency of X-ray induced mutations in Habrobracon eggs. Genetics 39, 851–856.

WOLFF, S. 1959. Interpretation of induced chromosome breakage and rejoining. Radiation Research Suppl. No. 1, 453–462.

WOOD, T. H. 1959. Inhibition of cell division. Radiation Research Suppl. No. 1, 332–346.

ZIRKLE, R. E., and BLOOM, W. 1953. Irradiation of parts of individual cells. Science *117*, 487–489.

ZIRKLE, R. E., and PARRISH, M. E. 1950. Comparative lethal action of X-rays and cyclotron neutrons on Drosophila eggs. U. S. Atomic Energy Commission Document No. CH-3409.

PART IV

Radiation Processing Technology

CHAPTER 15

The Nature of Food and the Role of Radiation

The Nature of Food

To survive and live effectively, man must engage in the eternal competition for food with all other forms of life. A plant is successful only if it can meet the limitations of soil, site, and climate in the presence of other plants and the usage of plants by animals. Some animals live directly on the plants, other animals demand flesh diets, and take it where they can. The flesh eating populations on earth must necessarily be the fewer in number. Man operates at the summit of a pyramid of life that throughout nature has as its base the vegetation of the earth. He can live and grow only when he has the proper amounts and kinds of nutrients. (Table 26 p. 188 and Table 40).

Men have been meat eaters from the beginning, when they had a choice (Jensen 1953). Actually, animal flesh closely resembles our own (Fig. 92). We find the balance of the amounts and kinds of nutrients that we need more nearly in meat, than in plant foods. Animal tissues all through nature have much in common. We can make fewer nutritional mistakes by eating fresh meat that we have cooked slightly, than by choosing from a hundred plants those nutrients we need. But we enjoy eating succulent fruits and vegetables; they add to the pleasure of eating and contribute importantly in essential nutrients, especially the water soluble vitamins.

The food harvests are oriented with the movement of the earth around the sun. Man's hunger wave is oriented with the spin of the earth. These two are not in harmony. The annual harvest of food per acre of land, under the best circumstances, in terms of man's daily requirement is summarized in Table 41. The yield of food per acre actually varies by a factor of perhaps ten, depending on the fertility of the soil, the application of known agricultural technology, and the climate and season patterns. An acre of Indiana's best farmland, given the climate and sunlight of the tropics might increase its productivity as much as four-fold, while in the environment of Alaska its productivity would drop perhaps to one half. The idealized productivity in the tropical climate is, however, much reduced by an abundance of crop disease and crop spoiling organisms. Up to the present, this has forced the main crop producing areas to be situated farther away from the equator. The disease problems are two-fold: they prevent the successful growth of a crop and they spoil the

TABLE 40

RECOMMENDED DAILY DIETARY ALLOWANCES[1]

	Calories	Protein Gm.	Calcium Gm.	Iron Mg.	Vitamin A, I.U.	Thiamin, Mg.	Riboflavin, Mg.	Niacin (Nicotinic acid) Mg.	Ascorbic acid, Mg.	Vitamin D, I.U.
Man (154 lbs., 70 kg.)										
Sedentary	2,400	70	1.0	12	5,000	1.2	1.8	12	75	
Physically active	3,000	70	1.0	12	5,000	1.5	1.8	15	75	
With heavy work	4,500	70	1.0	12	5,000	1.8	1.8	18	75	
Woman (123 lbs., 56 kg.)										
Sedentary	2,000	60	1.0	12	5,000	1.0	1.5	10	70	
Moderately active	2,400	60	1.0	12	5,000	1.2	1.5	12	70	
Very active	3,000	60	1.0	12	5,000	1.5	1.5	15	70	
Pregnancy (latter half)	2,400	85	1.5	15	6,000	1.5	2.5	15	100	400
Lactation	3,000	100	2.0	15	8,000	1.5	3.0	15	150	400
Children up to 12 yrs.										
Under 1 yr.	[2]	[3]	1.0	6	1,500	0.4	0.6	4	30	400
1–3 yrs. (27 lbs., 12 kg.)	1,200	40	1.0	7	2,000	0.6	0.9	6	35	400
4–6 yrs. (42 lbs., 19 kg.)	1,600	50	1.0	8	2,500	0.8	1.2	8	50	400
7–9 yrs. (58 lbs., 26 kg.)	2,000	60	1.0	10	3,500	1.0	1.5	10	60	400
10–12 yrs. (78 lbs., 35 kg.)	2,500	70	1.2	12	4,500	1.2	1.8	12	75	400
Children over 12 yrs.										
Girls 13–15 yrs. (108 lbs., 49 kg.)	2,600	80	1.3	15	5,000	1.3	2.0	13	80	400
Girls, 16–20 yrs. (122 lbs., 55 kg.)	2,400	75	1.0	15	5,000	1.2	1.8	12	80	400
Boys, 13–15 yrs. (108 lbs., 49 kg.)	3,200	85	1.4	15	5,000	1.5	2.0	15	90	400
Boys, 16–20 yrs. (141 lbs., 64 kg.)	3,800	100	1.4	15	6,000	1.7	2.5	17	100	400

[1] Courtesy of National Research Council.
[2] 110/2.2 lb. (1 kg.).
[3] 3.5/2.2 lb. (1 kg.).
From Desrosier (1959).

TABLE 41

PER ACRE PRODUCTION OF FOOD IN TERMS OF DAILY NUTRIENT REQUIREMENTS[1] OF MAN[2,3]

Type of Product	Value	Biological Unit	No. of Items	Food Energy (Days)	Protein (Days)	Fat (Days)	Minerals (Days)	Vitamins (Days)	Average for 10 Nutrients (Days)
Dairy and meat	Greatest	Whole milk	1	108	236	245	722	784	210
	Average		11	64	139	204	237	461	111
	Least	Sheep	1	11	23	39	28	87	19
Grains and staple food crops	Greatest	Soy bean, whole	1	521	2,224	1,078	4,240	4,796	1,286
	Average		17	653	673	197	1,353	1,933	481
	Least	Rye, flour	1	291	309	30	319	475	142
Vegetables, fresh	Greatest	Carrot	1	878	1,009	235	6,793	152,499	16.141
	Average		22	308	517	76	2,463	21,151	2,451
	Least	Sweet corn	1	187	274	84	286	2,591	342
Fruit, fresh	Greatest	Orange	1	769	582	131	3,421	35,768	4.067
	Average		13	493	267	286	1,346	12,294	1,469
	Least	Blueberry	1	92	34	32	348	1,388	189

---

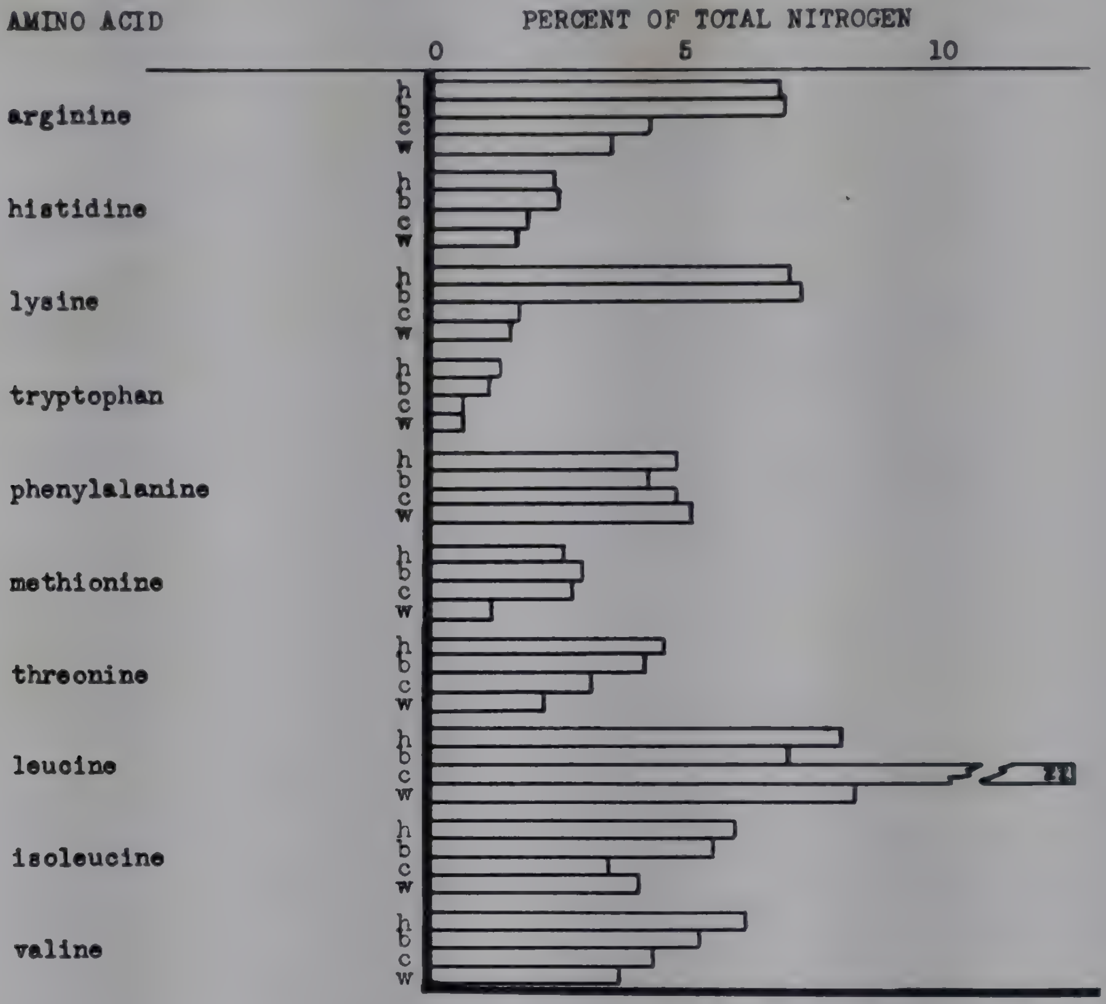
[1] The daily requirement of the National Research Council used in this table is: food energy 3,000 calories, protein 70 grams, calcium 800 mg., iron 12 mg., vitamin A 5,000 I.U., thiamin 1.5 mg., riboflavin 2.0 mg., and ascorbic acid 75 mg. No recommended allowance is given for fat, but it is considered desirable that 20 to 25 per cent of the food energy be in this form. This is the basis for the recommendation of 67 grams used above.

[2] Courtesy of MacGillivary.

[3] From Desrosier (1959).

crop after harvest in a very short time due to existing humidity and temperature conditions.

The amount of individual effort each of us can expend for our society and for mutual good is dependent on having more than the basic minimum of food for survival. Optimum human nutrition affords the foundation for a sustained optimum performance.



Courtesy of Mitchell and Block

FIG. 92. COMPARATIVE ESSENTIAL AMINO ACID COMPOSITION OF HUMAN, BEEF, CORN, AND WHEAT PROTEINS

We seek to maximize human conquests over natural destructive forces. If we look at the men who preceded us on earth, we see that the greatest developments of the human race, considering achievements, have been located in a few areas. These can be thought of as two zones bordered on one side by the frozen tundra and on the other by the disease infested tropics. The northern and southern hemispheres offer equally good opportunities for advancement. We are able to extend these belts towards the tropics with modern sanitation and technology. Those regions on earth where civilizations occurred at an early date were areas where food production and food storage from one harvest to the next were possible because the climate was favorable for such activity. The preservation and storage of foods was an important factor involved in the civilization of man. Continued improvement in this food technology is required to keep abreast of population increases and population concentrations.

The increase in dense urban human populations has been underway since before the Middle Ages. This has posed and continues to pose critical problems of supplying large human populations with their daily foods. For example, there are more people currently on the island of Manhattan in New York City than there were on the entire continent of North America at the time of Columbus. Consider the magnitude of the daily food supply problem.

The food supply has been an important consideration in the development of civilization. As populations increased and concentrated in various geographic areas, urban societies developed. The necessity of feeding a population removed from the food production site forced developments in food preservation. The principal methods in use currently are drying, salting, pickling, cold storage and frozen storage, canning, and chemical preservation.

Until the advent of radiation, no new method of preserving foods has appeared since the discovery of canning in the early 19th century. Technological developments have resulted in many improvements in food preservation methods so that quality of foods so processed has been greatly improved. However, the conventional methods of food preservation produce undesirable changes in many foods, including inactivations of vitamins, discolorations, off-flavors, denaturation and coagulation of proteins. Freezing for instance also requires a continuous application of the process, or the food decomposes. Drying has disadvantages in the acceptability of the reconstituted foods, and pickled products are but temporarily preserved. Chemical additives in part satisfy the requirement of heatless sterilization, however, the additive itself, by its nature, in most instances is objectionable as an addition to the diet. Radiation may enjoy a special position in preserving, disinfecting, and stabilizing man's food.

Yet, we see the food distribution problem as two sided. One, it is necessary to supply an adequate diet in the biological sense. Two, a satisfactory diet must also be supplied in the cultural sense. Depending on the requirements of a particular culture, the diet should prevent the problem of monotony, as it is directly related to morale, enjoyment of life, and our contribution to society. Food preferences exist in the various geographical areas of earth, and are successful solutions evolved from local food supplies, the useful information being perpetuated in the culture.

A list (Table 42) of 22 foods is available which could offer a minimum long range satisfying diet for man. The list is composed of important meats, poultry, fish, fruits, vegetables, and grain. Obvious substitutions are apparent and are made daily. Nevertheless, the foods on this list can be arranged into meals, and yield feeding patterns which will not be monotonous, providing they are available in acceptable quality.

TABLE 42

TABLE OF TWENTY-TWO FOODS

Meat	
	Beef, chicken, lamb, pork, ham, bacon
Fish	
	cod, shrimp
Fruits	
	peaches, lemons, oranges, dried fruit, fresh berries
Leafy and green vegetables	
	broccoli, cabbage, green beans
Root crops	
	white potatoes, sweet potatoes, carrots
Cereals	
	flour, whole grains
Milk	
	cow

A man consumes about 10 times his body weight in food annually; the people on earth require annually about 40 trillion pounds of food. While this much food is available on earth in one place or another, a goodly portion never benefits mankind, and most human beings are malnourished. The food problem is not solely in food production; it is equally one of food distribution.

All foods are perishable commodities; they begin to deteriorate shortly after harvest, gather, or slaughter. Some deterioration is accompanied by the production of poisonous agents; other deteriorations inflict losses in the nutritive value of foods. And, food is a main avenue in the spread of parasitic infections of man.

Radiation technology has much to contribute to the solution of that seemingly ageless problem of an adequate food supply for all men. The successful application of present knowledge could go far in improving the lot of mankind.

Safe Food for Man

Although food preservation in one form or another has been practiced throughout the history of man, it was not until Louis Pasteur that an explanation was found as to why foods spoil. The last word has yet to be written.

There is no agreement in the world as to what constitutes spoiled food. What is considered inedible to one group of people (certain cheeses) may be highly desirable to another group. Eggs are considered edible by Orientals when they are partly decomposed. Others consider such items spoiled and unfit for consumption by humans. In any event, all agree that spoiled food should not be eaten! Fortunately, many deteriorated foods will not injure our health, but the nutritive and aesthetic values of the food may be altered. It is generally difficult to identify by appearance which deteriorated foods will cause us illness.

Micro-organisms are present in our foods, in air, soils, dust, and water, and we carry them ourselves. All they usually need to start their process is a source of energy, such as a few drops of juice containing a trace of sugar. If the food is moist and the organisms have humid growing conditions, the invasion is on.

In order to feed ourselves we must act to deter these natural processes. Decomposition is a natural phenomenon of our foods; it is an inescapable problem. Living tissue has resistance to the degradation action of micro-organisms. Plant or animal tissue that is dead is consumed by biological forces in one way or another. There is a contest between man, the animals, and bacteria, yeasts, and molds as to which will consume the nutrients first.

In attempting to prevent the deterioration of plant and animal tissues, man has a doubly difficult task in that he must not only preserve the food for his own use but he must also exclude the other forces of nature from becoming active.

The principal causes of spoilage in foods are the growth of micro-organisms, the action of naturally occurring enzymes in the foods, chemical reactions, and physical degradation and desiccation. The type of spoilage for a particular food item depends to a great extent on the composition, structure, types of micro-organisms present, and conditions of storage of the food.

Micro-organisms have nutrient and environmental requirements, as do other plants. Specific food spoilage is generally associated with certain types of foods and micro-organisms. Some of the factors which control the type and extent of microbial food spoilage are the moisture content, the temperature, the oxygen concentration, the nutrients available, the degree of contamination with spoilage organisms, and the presence of growth inhibitors. Generally the control of one or more of these factors controls microbial spoilage.

Cleanliness is obviously one of the most important deterrents of food spoilage. Microbiological deterioration of a food involves multiplication and growth of billions of cells within a day or two following initiation of growth. It follows then that whatever we can do to reduce the numbers of micro-organisms will prolong the useful life of the food. The larger the initial population of organisms, the more rapidly the food is attacked. The application of good sanitary practices in the handling of foods does reduce the incidence of spoilage.

Time is another important consideration. The growth of spoilage organisms is a function of time as well as temperature and environment. The reduction of the amount of time elapsed between harvest and consumption is another important consideration in the control of food spoilage.

With the exception of chemical poisons and parasitic infections, foods which we harvest or gather or slaughter and consume without storage offer little danger to us when properly prepared for consumption.

Food infections and food intoxications may occur when we store or prepare or preserve our food. It takes a combination of time and proper environment for microbial spoilage to occur (Dack 1956). Unfortunately, it is not possible to mass distribute or mass produce our food so that this combination does not exist. We must therefore prevent deterioration in our foodstuffs (see Table 43).

TABLE 43

USEFUL STORAGE LIFE OF FRESH UNTREATED PLANT AND ANIMAL TISSUES AT VARIOUS TEMPERATURES

Food	Estimated Average Useful Storage Life (Days) at		
	32°F.	72°F.	100°F.
Animal flesh	10 to 60	1	Less than 1
Fish	5 to 30	1	Less than 1
Poultry	7 to 21	1	Less than 1
Fruits	2 to 180	1 to 20	1 to 7
Leafy vegetables	3 to 30	1 to 7	1
Root crops	90 to 300	7 to 50	2 to 10

Potatoes, carrots, beets, cabbage, and turnips can be stored alive. These foods left alone in nature may live through the winter and bear seeds the second year. Cabbage may remain alive so long as sugar is left in its leaves for energy; such plant tissues killed by heating or freezing lose their ability to resist invasion by micro-organisms.

Fruits, although alive, differ from the vegetables in one important aspect. Fruit cells are rather old cells, incapable of reproducing themselves. They are not destined by nature to grow, but to deteriorate and supply an appropriate seed bed for the germination of the seed. Fruits are not resistant to injury. While a potato can heal a wound, a mature fruit usually can not—an important difference.

The decomposition of dead plant and animal matter is not a simple process, but represents the combined effort of many different types of micro-organisms.

The pleasure we obtain from food is dissipated when the food is decayed. Our ancestors may not have been easier to please, they just had no choice.

Food poisoning has been with us from the beginning (Table 44). Cases of food poisoning are relatively numerous today and probably have been even more common in the past. Whenever possible, we should eat sterile foods, keep our perishable meat foods cold, consume live and healthy plant foods, or plant and animal foods that are properly preserved by one method or another. If we give close attention to cleanliness of eating utensils and keep insects and other pests from our tables and our foods, we leave little opportunity for food poisoning to occur. We have learned not to keep foods in any form which are prepared and perishable (moist and warm) without refrigeration.

TABLE 44

CHARACTERISTICS OF FOOD POISONING CAUSED BY BACTERIA[1]

Symptoms	Start of Symptoms After Eating	Type Poisoning	Cause Eating of Toxin	 Eating of Organisms	Mortality Per cent
Vomiting, nausea, diarrhea, abdominal cramps	1–6 hours or less	Staphylococcus	Yes	Yes	1 or less
Vomiting, nausea, chills and fever, diarrhea	7–72 hours	Salmonella	No	Yes	1 or less
Nausea, colicky pains, diarrhea	5–18 hours	Streptococcus	No	Yes	1 or less
Difficulty swallowing, double vision, difficulty breathing, paralysis	1–2 days	Botulism	Yes	Yes	70 or more

[1] Adapted from Dack (1956).

Asepsis is found naturally occurring. Starches are packed in sealed packages (grains) which are not broken by micro-organisms under ordinary conditions. Fruits and vegetables are covered with an impervious skin which resists microbial invasion. Except for surfaces exposed to air and dirt, internal, healthy plant and animal tissues are sterile.

As a general index for growth of mold, yeast, and bacteria, the following may be useful:

Mold will grow in a food as dry as grain if air is present and warm humid conditions prevail.

Yeasts need slightly more water than molds, but can grow in air or without air in warm humid conditions.

Bacteria also need more moisture than molds but can grow in air or without air in warm humid conditions. Temperatures near the freezing point of water restrict the growth of all organisms. Hence, cold temperatures have been our simplest control over microbial growth.

Foods which are free of micro-organisms and kept so will not undergo microbial spoilage, but some of them may be susceptible to chemical deterioration. Sterile foods are subject to chemical reactions which may cause losses in flavor, color, and texture.

Food Groups

It is useful to classify the foods of man on the basis of acidity. Plant tissue (except fruits and berries) and animal tissue (including meat, fish, and dairy products) are classed as low acid foods, with pH values ranging from 5.0 to 7.2. Manufactured items with several ingredients may fall into the medium acid group with pH values ranging from 4.5 to 5.5. Fruits are in the medium acid group, ranging in pH from 3.7 to 4.5. Berries, fermented products, and certain citrus products fall into the high acid group, with pH values below 4.0, as are jams and jellies. Except for eggs (pH 7.5), few foods are basic in reaction if in the best condition.

Micro-organisms Associated with Food Groups

Bacteria can be classified according to their temperature requirements for growth. Bacteria of the soil, water, air, and body growing at room temperature or slightly higher are called mesophiles; their range is between 70°F. and 110°F. Some water and soil type bacteria grow best at temperatures ranging from 35°F. to 50°F., and are called psychrophiles. There are bacteria of soil, water, and air which grow best at temperatures from 120°F. to 170°F., and are called thermophiles.

It is important to distinguish between two groups of organisms, those capable of growing at moderately high temperature, the thermophiles, and those incapable of growing at these temperatures yet surviving the effects of the temperature, the thermoduric group. The mesophiles of importance in food spoilage in their spore stage are thermoduric.

Foods have associated microfloras; certain organisms are associated with particular food groups. These organisms gain entrance to the food during processing either from the soil, from ingredients, or from equipment. On the basis of the acidity classification of foods, it is possible to make general statements relative to the spoilage organisms encountered which are of importance to the success of the preservation or spoilage processes (Tanner 1944).

Typical genera of micro-organisms commonly associated with the spoilage of important fresh foods are given in Table 45.

TABLE 45

COMMON SPOILAGE ORGANISMS OF FOODS

Food	Organisms Commonly Found in Spoiled Food
Milk and milk products	*Streptococcus*, *Lactobacillus*, *Microbacterium*, *Achromobacter*, *Pseudomonas* and *Flavobacterium*, *Bacillus*
Fresh meat	*Achromobacter*, *Pseudomonas* and *Flavobacterium*, *Micrococcus*, *Cladosporium*, *Thamnidium*
Poultry	*Achromobacter*, *Pseudomonas* and *Flavobacterium*, *Micrococcus*, *Penicillium*
Smoked cured meats	*Micrococcus*, *Lactobacillus*, *Streptococcus*, *Debaryomyces*, *Penicillium*
Fish, shrimps	*Achromobacter*, *Pseudomonas* and *Flavobacterium*, *Micrococcus*
Shellfish	*Achromobacter*, *Pseudomonas* and *Flavobacterium*, *Micrococcus*
Eggs	*Pseudomonas*, *Cladosporium*, *Penicillium*, *Sporotrichum*
Vegetables	*Penicillium*, *Rhizopus*, *Lactobacillus*, *Bacillus*, *Achromobacter*, *Pseudomonas* and *Flavobacterium*
Fruits and Juices	*Saccharomyces*, *Torulopis*, *Botrytis*, *Penicillium*, *Rhizopus*, *Acetobacter*, *Lactobacillus*

Botulism.—From a public health standpoint, for permanent preservation, foods with pH values greater than 4.5 must be processed so that the sterilization of *Clostridium botulinum* is effected. The lower limit of growth of this important food poisoning organism is at a pH value of 4.5. Inasmuch as a millionth of a gram of the toxin produced by this organism will kill man, certain precautions are indicated. Due to the public health significance of *Clostridium botulinum*, it is worthwhile to discuss this organism briefly.

Many people at one time or other have eaten this bacterium, as it is a common soil organism. There are at least four types, two of which (A and B) are noteworthy here. Type A occurs generally only in virgin soil or newly reclaimed forest areas, type B in cultivated soils.

This organism is a gram positive, anaerobic, spore-forming bacterium. It develops in the absence of atmospheric oxygen, or in very low oxygen tension environments.

The organism exists in both a vegetative form and in the form of a radiation resistant spore. While the vegetative form is killed by less than 1,000,000 rads, some spores on the other hand may survive more than 4,000,000 rads. The *D* value for these spores was discussed earlier and is 400,000 rads. The spores are the important form of the organism from the radiation processing viewpoint, spores being found in dust and soil. The main contaminations are either from the soil in which food is grown or infected animals (limberneck in chickens). Spores of *Cl. botulinum* have been isolated from most of the earth's surface. It is a difficult organism to work with because it loses its toxin producing capacity when manipulated in laboratory media. Spores vary in radiation resistance, and it is not simple to obtain a spore suspension of uniform resistance for study.

The organism has both proteolytic and saccharolytic powers. The medium in which a culture is growing actively has an odor of a composite of decomposed meat mixed with butyric acid. However, it is not unusual to find the organism present, with its toxin, and still have the material reported as not objectionable to taste or smell. Hence, persons ingesting such food may not suspect the food which poisoned them. *Cl. botulinum* has a heat resistance at 250°F. of 2.8 minutes for 10,000 spores per ml. in neutral phosphate buffer solution, hence this problem is significant in the canning of food or even in freezing, due to dangers during thawing.

The botulina toxin produced is water soluble, and extremely lethal to man. It has been characterized chemically recently by research workers.

The spores must germinate in order to produce a vegetative cell to produce the toxin. The toxin is destroyed by a ten minute exposure to boiling water. Heating foods in preparation for eating is a valuable process.

A suitable anaerobic condition for this organism to grow would be a sealed can with a vacuum, or deep in a liquid, or in the center of inoculated meat products, or in oil packed fish, etc. However, aerobic organisms may grow and deplete oxygen in a container, creating anaerobic conditions adequate for growth of *Cl. botulinum*. In an acid food, when the acid has been utilized by other (mold) organisms, raising the pH, *Cl. botulinum* may grow if present. All foods capable of sustaining the growth of this organism should be sterilized on the assumption that the organism is present and must be destroyed.

Origin of Spoilage Organisms.—Troublesome spoilage micro-organisms as noted above are organisms of soil, water, air, plants, and animals.

Organisms of Soil.—While all the spoilage types are to be found in the soil, unless there is a transfer of soil (or dust) to the container of food, soil is not an important source of spoilage organisms. There is, of course, the contamination of raw products taken from the soil, but ordinary processing should find soil washed adequately from food.

The surface of certain foods not peeled nor prepared will contain varying numbers of soil borne micro-organisms. Asparagus and green beans could be contaminated with soil unless the washing process is adequate. Meats are prepared from butchered animals, and soil contamination would be unlikely. Certain animal products such as pig's feet may be an exception, not different from asparagus relative to soil inoculation. Dust carried through air to food products may be a source of contamination.

Equipment.—Spoilage micro-organisms can normally be expected to be present on equipment, particularly that having a steam connection due to the creation of favorable temperatures in equipment for growth of organisms. Only microbiologically, physically, and chemically clean equipment can give desired results. Contamination of foods by equipment means either poor sanitary practices, poor equipment design, poorly utilized equipment, or a combination of these factors. It is not possible to clean equipment constantly during operating periods, and in spite of a sanitation program, it is possible to have clean equipment contaminated by the flow of raw food products.

Equipment is a constant source of contamination of food products in factories. Careful attention to sanitation practices and the condition of raw products flowing through a plant is essential to successful radiation preservation of foods. Radiation destruction of organisms was shown earlier to be dependent on the concentration of organisms present.

Food Ingredients.—Common ingredients in foods have been found to be sources of mesophilic and thermophilic spore-forming bacteria as well as other micro-organisms. The standards established for these ingredients by the canning industry are useful. Other ingredients including dry milk solids, syrups, flours, and spices are generally contaminated with micro-organisms. Using sugar having only 40 spores per gram when made into the packing solution could yield 500 or more spores per package. Hence, careful attention to ingredients which are added to foods during preparation and preservation, is indicated.

Radiation Destruction of Food Spoilage Organisms

Micro-organisms in Low Acid Foods.—In foods with a pH value greater than 4.5, mesophilic spore-forming anaerobic bacteria are important, *Clostridium botulinum* being an example. Another is one known as Putrefactive Anaerobe (P. A.) No. 3679, a *Clostridium sporogenes* type, common in soil. The latter is less radiation resistant than the former, the former being used to evaluate radiation processing schedules. Sufficient irradiation to kill the spores of *Clostridium botulinum* in a process insures the destruction also of P. A. No. 3679.

In addition to the mesophilic spore forming bacteria there are also thermophilic spore-forming organisms, which also are very resistant. These organisms therefore are controlled through sanitation and by strict control of ingredients.

Micro-organisms in Acid Foods.—In the acid food group, the troublesome organisms are aciduric bacteria of no special radiation resistant qualities. Bacteria, yeasts, and molds are capable of spoiling these foods. The lack of growth of *Cl. botulinum* in acid foods is reflected in their low processing requirements (D value of 200,000 rads).

Thermophilic flat sour spoilage is due to an inoculation of food usually by equipment or ingredients and is related to the sanitary conditions of a plant. In flat sour spoilage, as the name implies, acid is produced without gas. One difficulty then is that containers do not appear to be spoiled until opened.

It is a general axiom of the food industry never to taste spoiled low or medium acid foods, due to the threat of spoilage by *Cl. botulinum.* With acid and high acid foods, the spoiled products may be distasteful, but there is, as noted above, reasonably little danger in tasting them. Fortunately, most food spoilage is associated with the production of odors, and bulging of the container. There are usually visible signs of decomposition in the food iteslf.

Micro-organisms in High-Acid Foods.—Aciduric bacteria, yeasts, and molds are the troublesome organisms in this group. Their radiation resistance is generally low, a *D* value of less than 200,000 rads. In this group, too, the natural enzymes present in food are more radiation-resistant than micro-organisms. Due consideration must be given to the destruction of natural enzymes in foods of all groups for permanent preservation.

To summarize the above, foods with pH values greater than 4.5 require relatively severe radiation treatments for sterilization. Bacterial spores are more resistant to the germicidal action of ionizing radiations than are vegetative cells. *Cl. botulinum* type A spores are the most radiation resistant as well as the most dangerous of the food spoilage bacteria (*D* value of 400,000 rads). The safe sterilizing dose requirements for *Clostridium botulinum* in foods with pH values higher than 4.5 is 4.8 million rads, on the basis of current knowledge.

Foods of high acidity can be sterilized with radiation doses near 2.5 million rads. Yeasts and molds are generally killed with doses in the 500,000 rad range. Molds on fruits are killed with doses in the 100,000–200,000 rad range.

It should be recalled that the concentration of organisms influences the number of survivors in a food. The order of death to micro-organisms in foods subjected to ionizing radiations is logarithmic. The killing effects of ionizing radiations can be improved by increasing the temperature of the food during irradiation.

Most micro-organisms if actively growing (vegetative state) are readily killed by exposure to temperatures near the boiling point of water, hence are easily controlled in radiation processing, which generally requires enzyme inactivation by moist heat for permanent preservation. This will be discussed later.

Role of Radiation

There are at least six distinct areas of application for radiation processing of foods. First, there is permanent preservation. To qualify, a radiation-sterilized product must conform to high standards of consumer appeal, nutritive value, wholesomeness, economy, and storage stability in competition with other preserved products. Second, the application of limited radiation doses is useful to prolong the storage life of market commodities such as cut meats, fresh fish and fresh fruits and vegetables with or without refrigeration. The satisfactory extension of storage life of natural commodities without marked losses in quality characteristics has yet to be solved, and radiation may enjoy a special position in this area.

Third, the destruction of insects in various life cycle stages in food products is feasible with ionizing radiations. This can be accomplished completely. Fourth, growth processes of plant tissues are radiation-sensitive. Examples of the application are seen in sprout inhibition of potatoes and onions. Fifth, ionizing radiations have promise as a unit operation in the food industries, i.e., preparation of sterile enzyme solutions, hydrolysis of large molecules such as cellulose, tenderization of meat, improved roasting methods for coffee beans, and aging of wines. Sixth, the destruction of parasites in man's foods, the desensitization of proteins, and the control of food poisoning organisms in foods can be accomplished by radiation.

Man's food supply is composed of plant and animal tissues which have individual components in common. These components are identifiable and their specific interaction with ionizing radiations has been discussed earlier. In the following three chapters the influence of radiation on whole food substrates and packages will be discussed.

Physiological Aging of Plant Tissues and Organs

As indicated in an earlier chapter, the metabolic processes continue in an intact plant organ even after separation of the organ from the parent plant. There are physiological activities in harvested fruits and other plant tissues, and, even in the absence of micro-organisms or spoilage agents, the tissues continue to respire and age. The process is enzymatically controlled and proceeds uninterrupted in the tissues. There are therefore limits to the storage life of sterile but living tissues. For example, after 20 days of storage under ideal refrigeration conditions, sterile strawberries physiologically break down. Similar breakdown occurs with all plant tissues in time. For permanent preservations, the physiological processes must be halted, e.g., enzyme systems must be inactivated.

Autolysis of Animal Tissues

With the death of animals, an almost immediate degradation of tissue occurs. Witness the softening of the belly of a freshly caught trout in a matter of a few hours following catching. Even with sterile flesh, this self digestion or autolysis occurs. It is necessary to halt these autolytic processes if the animal tissue is to be retained for some period of time greater than a few days even with refrigerated storage. For permanent preservation, the enzymatic systems present in animal flesh must be inactivated even in sterile tissues. The subsidence of rigor mortis is due to autolysis, and this is usefully applied in the controlled aging or tenderization processes for meat animal carcasses.

BIBLIOGRAPHY

Blanck, F. C. 1955. Handbook of Food and Agriculture. Reinhold Publ. Co., New York.

Braidwood, R. J., Krogman, W., and Tax, S. 1946. Time, Space, and Man. University of Chicago Press, Chicago, Ill.

Brownell, L. E. 1958. Utilization of Gross Fission Products. Engineering Research Institute, University of Michigan, Ann Arbor, Michigan.

Childe, V. G. 1946. Man Makes Himself. Watts and Company, London, England.

Clifcorn, L. E. 1955. The food industry's attitude toward radiation sterilization. Nucleonics *13,* No. 1, 39–41.

Cook, R. C. 1951. Human Fertility: The Modern Dilemma. William Sloane Associates, New York.

Cook, R. C. 1958. World population summary. Pop. Bull. *13*, 133–151.

Cook, R. C. 1958. Population inflation shrinks the earth. Pop. Bull. *4*, 62–83.

Dack, G. M. 1956. Food Poisoning. University of Chicago Press, Chicago, Ill.

Desrosier, N. W. 1958. Food irradiation research and its practical application in Europe and the United States. Project Report No. 396. The European Productivity Agency, Organization for European Economic Cooperation, 2 Rue Andre Pascal, Paris, France.

Desrosier, N. W. 1959. A challenge to the food industry of the U. S. A. in the peaceful uses of atomic energy. Food Processing *19*, 30, 106–109.

Desrosier, N. W. 1959. The Technology of Food Preservation. Avi Publ. Co., Westport, Conn.

Frobisher, M. 1949. Fundamentals of Bacteriology. Fourth Ed. W. B. Saunders Co. Philadelphia, Penn.

Hannan, R. S. 1956. Preservation of food with ionizing radiations. Science and Technology of Food Preservation by Ionizing Radiations. Chemical Publ. Co., New York.

Jacobs, M. B. 1951. The Chemistry and Technology of Food and Food Products. Interscience Publishers, Inc., New York.

Jensen, L. B. 1949. Meat and Meat Foods. The Ronald Press Co., New York.

Jensen, L. B. 1953. Man's Food. Garrard Press, Champaign, Ill.

MacGillivray, J. H. 1956. Factors affecting the world's food supplies. World Crops *8*, 303–305.

Morgan, B. H. 1955. Food radiation round up. Food Eng. *16*, 43–47.

Orr, J. B. 1943. Food and People. Pilot Press, London, England.

Oser, B. L. 1958. The food industry in a changing world. Food Technol. *12*, 493–496.

Progress Report on Atomic Energy Research. 1956. Hearings before the Committee on Research and Development of the Joint Committee on Atomic Energy, Congress of the United States. U. S. Govt. Printing Office, Washington, D. C.

RUSSEL, E. J. 1953. World Population and World Food Supplies. George Allen and Uniwin, London, England.

SAX, K. 1955. Standing Room Only. Beacon Press, Boston, Mass.

SEBRELL, W. H., JR. 1953. Chemistry and the life span. Chem. Eng. News *27*, 3624–3629.

SHERMAN, H. C. 1946. Chemistry of Food and Nutrition. Macmillan Company, New York.

SILOW, R. A. 1958. The potential contribution of atomic energy to development in agriculture and related industries. Int. J. Applied Radiation & Isotopes *3*, 257–280.

STEFANSSON, V. 1937. Food of the ancient and modern Stone Age man. J. Am Dietet. Assoc. *13*, 102–119.

TANNER, F. W. 1953. Food Borne Infections and Intoxications. Twin City Printing Company, Champaign, Ill.

TANNER, F. W. 1944. The Microbiology of Foods. Garrard Press, Champaign, Illinois.

THIMANN, K. V. 1955. The Life of Bacteria. Macmillan Company, New York.

TISCHER, R. G. 1958. Feeding men during space flights. Food Technol. *12*, 429–430.

UNITED NATIONS. 1958. European Meeting on the Use of Ionizing Radiations for Food Preservation. Atomic Energy Research Establishment, Harwell, England. November 17, 1958.

UNITED STATES ARMY QM. CORPS. 1957. Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

VAUGHAN, W. T. 1940. Why we eat and what we eat. Sci. Monthly *50*, 148–154.

WELLS, H., and WELLS, P. H. 1956. General Biology. McGraw-Hill Book Co., Inc., New York.

ZCHEILE, F. P. 1950. Role of genetics in food quality improvement. Nutrition Rev. *8*, 65–69.

ZEIGLER, P. T. 1943. The Meat We Eat. Interstate Publishers and Printers, Danville, Ill.

CHAPTER 16

Wholesomeness and Acceptance of Irradiated Food

Recognizing the unique ability of ionizing radiation to create highly reactive chemical species in its path, and also the ability of certain high energy particles and photons to induce artificial radioactivity in elements, the question arises whether radiation treated foodstuffs are fit for human consumption. With the ever increasing availability of radioactive sources and their potential application in food stabilization, such food could conceivably constitute a major portion of the foods in the dietary. Initial investigations indicated noticeable alterations in odor, taste, color, and texture of exposed food. Chemical studies of these changes indicate that the substances responsible were present only in trace amounts, yet their detection with the senses was real. It is possible therefore that in addition to such changes which occurred, toxic products might also be produced in minute but significant amounts. To date, extensive laboratory tests have not revealed any manifestation of such toxic substances. In fact, evidence to date indicates irradiated foods are wholesome for man to consume in his diet (McNamara 1958).

Under the Miller Amendment of the Federal Food, Drug, and Cosmetic Act, any new food processing technique prior to its introduction in the U. S. A. requires a comprehensive and convincing body of evidence on the safety of such a process. Realizing the responsibility of the Food and Drug Commissioner, who administers the Act, conservatism on the question of clearance of radiation treated foodstuffs is essential, as is true of any new process. Furthermore it is imperative that conclusive proof be obtained of the absence of deleterious changes in irradiated foods and of the acceptability of treated foods in human diets. Two studies are clearly indicated, one dwelling on the potential toxicity of the treated foods, the other investigating the nutritional adequacy of the foods when introduced into the dietary.

In the U. S. A., the Food and Drug Administration has jurisdiction over such problems as relating to civilian populations. On the other hand, the Surgeons General of the Armed Forces have this responsibility for the military forces. In this instance this authority was delegated to The Surgeon General of the U. S. Army.

The Congress recognized that the task at hand to prove the usefulness of irradiated foods for man was of such magnitude that until satisfactory evidence could be obtained, the risk involved by any corporate organization was so great that it could not be undertaken privately. The problem required the attention of the Federal Government if this peaceful use of atomic energy was to be realized and yield its benefits to mankind. The funds to undertake the program were made available by the United States government, and the most elaborate experiment with food ever undertaken in the history of mankind was initiated to determine whether radiation stabilized foods were either useful or harmful.

Wholesomeness Studies

A test program was developed which would yield information with which judgment of irradiated foods would be made. For this effort more than 40 medical and nutritional research laboratories have participated. The tests were developed by the staff of the Office of The Surgeon General, and the National Academy of Science in cooperation with the Food and Drug Administration. Essentially the potential toxicity and nutritional adequacy of the foods were put to rigid evaluation. Considerations of freedom from carcinogenicity, antigenicity, and induced radioactivity were built into the testing procedures. Chemical and physical tests preceded animal tests in order to give insight into the problems which would be encountered and permit investigators to plan experimental programs adequately. To disclose toxic effects, animals were challenged with an excess of substances under consideration. For nutritional adequacy the usual supplements to the diets were reduced to minimal levels to aggravate any inadequacy. In addition to studies with normal, healthy animals, additional tests were conducted with animals under various conditions of stress, attempting to increase their susceptibility to toxic substance, if present. Recognizing the variations between species of animals, and differences in strains of animals, several species were used. In all tests it was pointedly marked that changes, if they should appear, would be subtle, based on preliminary evidence. Metabolic studies were devised, including cellular studies, to supplement gross observations relating to growth, reproduction, longevity, and behavior. Enzyme studies in tissues of sacrificed animals were included.

In addition, it was recognized that the effects of irradiation would be dose-dependent, and the test plan demanded that levels higher than those required for treating food should be included.

In sum total, the testing program took the following characteristics. Four animal species were selected—rats, chickens, dogs, and monkeys. After clearance by these animal tests, human volunteers were obtained to consume diets containing mainly irradiated foods. After this information was collected, studies could be undertaken with large numbers of human volunteer consumers, but now testing the acceptability of the treated food, and not wholesomeness, two quite distinct concepts (Fig. 93). In wholesomeness, it is considered whether or not the food can be consumed, and if consumed, if it does harm to the individual. In acceptance, the concept being tested was, now that the food had been shown to be useful in the dietary, will a consumer eat the food, and if he eats it, will he like it?

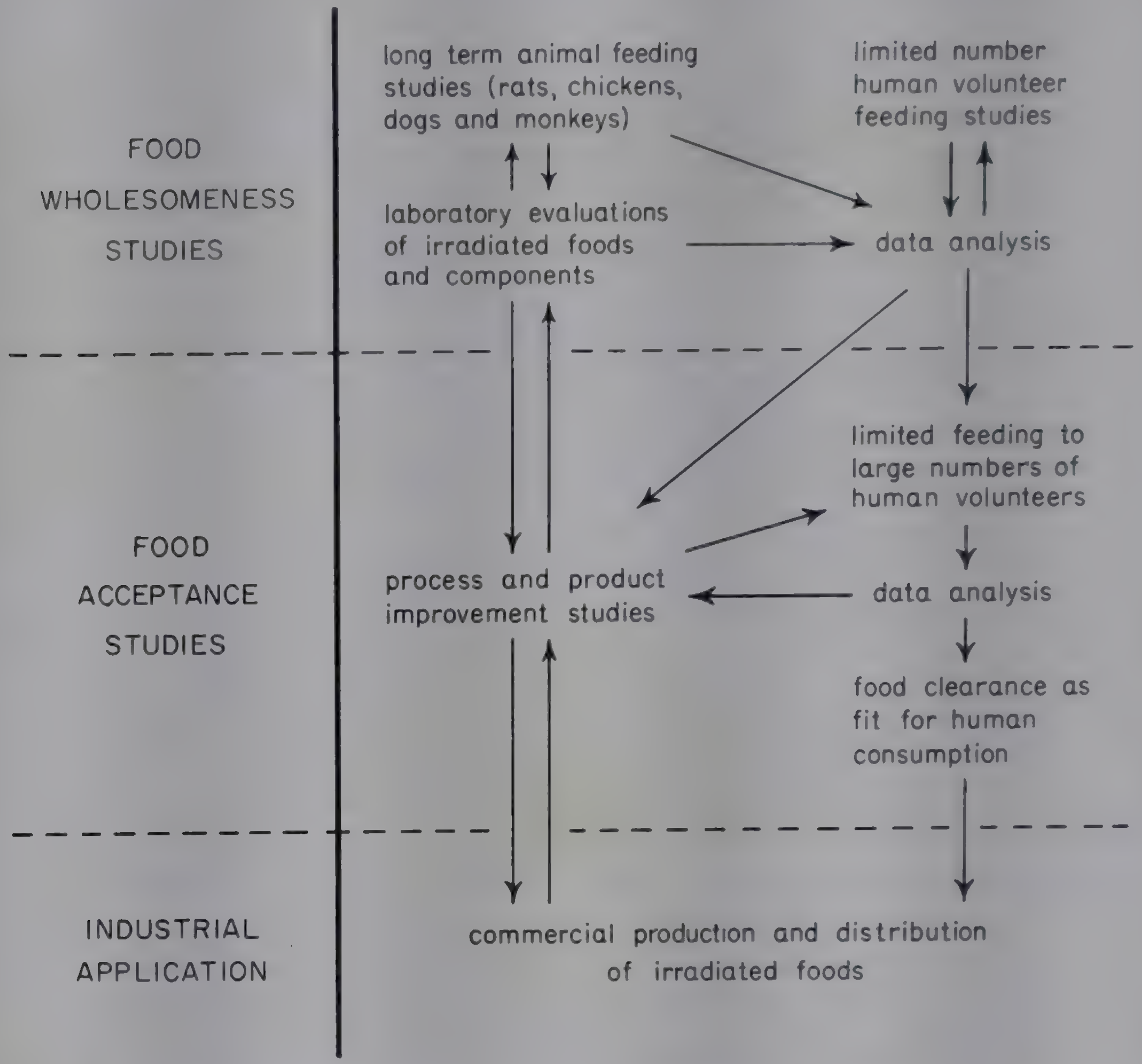

FOOD WHOLESOMENESS STUDIES
FOOD ACCEPTANCE STUDIES
INDUSTRIAL APPLICATION
long term animal feeding studies (rats, chickens, dogs and monkeys)
limited number human volunteer feeding studies
laboratory evaluations of irradiated foods and components
data analysis
limited feeding to large numbers of human volunteers
process and product improvement studies
data analysis
food clearance as fit for human consumption
commercial production and distribution of irradiated foods

FIG. 93. SCHEMATIC DIAGRAM OF TEST PROGRAM FOR ESTABLISHING WHOLESOMENESS AND ACCEPTANCE OF IRRADIATED FOODS FOR INDUSTRIAL APPLICATION

The test programs were of two years duration, with each animal species. More than 250,000 animals have participated, through several generations. This program started in 1954, and should culminate in 1960. Foods were treated at 0, 3×10^6, and 6×10^6 rad levels, to yield information on dose levels higher than thought useful, to accentuate deviations from normal if they were to appear.

Initially foods were irradiated in a frozen condition, and stored frozen, to demonstrate solely the influence of irradiation on the effects which might appear. It was shown with these studies on some 100 odd foods that there were no detectable harmful effects from eating the foods. Then, with this background, and recognizing the difficulties of operating a test with more than 100 items, a reduced list of 22 foods was evolved, which, if successfully treated, could be extrapolated to include most of the 100. These important classes were composed of meats, fish, fruits, vegetables, poultry, and cereal products (see Table 42 p. 288). Furthermore, studies were to be conducted with food stored at room temperature, at least three months old. Eight foods have progressed at this writing (1959) through the entire test program to the point of being allowed in large scale acceptance tests. The remaining 14 foods are at various stages of proceeding through the program. Concerning the program of wholesomeness, the relevant information presented in earlier chapters can be summarized as follows:

–proteins undergo alterations, and while chemical changes may be induced, nutritional adequacy is still acceptable, and toxic products have not been found (Table 46).

–lipid components of foods may undergo peroxide formation, but this is not objectionable. In natural fats, peroxides are initially relatively high but decrease on storage; no toxic products have been uncovered.

–carbohydrates undergo molecular alterations, mainly a degradation to smaller units, which in some cases makes them more available in metabolism, and no toxic products have been uncovered.

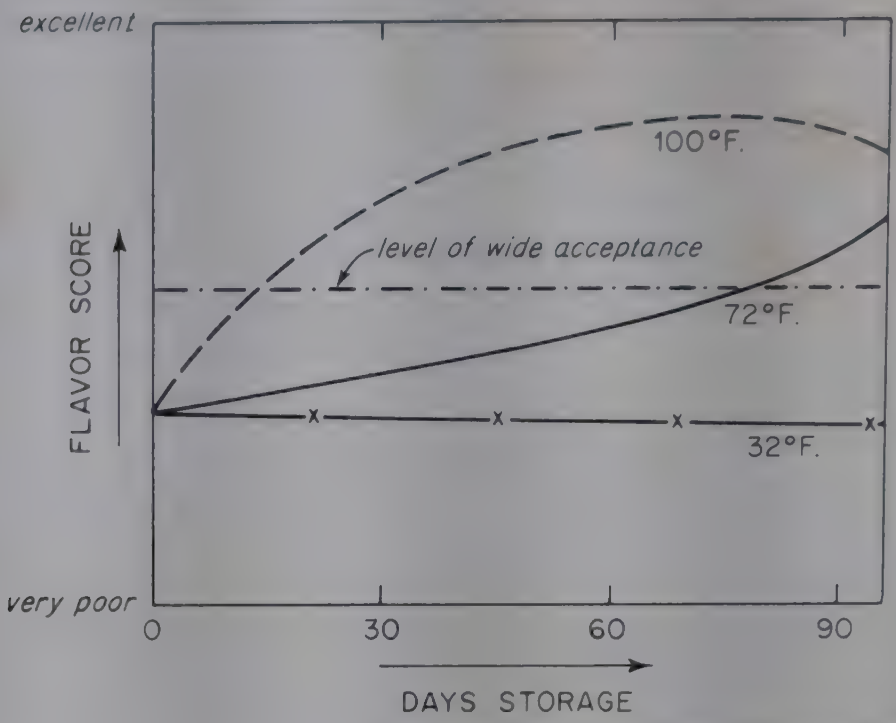
–of the trace nutrients, vitamin K has been found to be radiation-sensitive, and this can be of significance to the rat, although for humans of very minor significance, and the difficulty can be overcome by the addition of the vitamin. Other vitamins may undergo various degrees of destruction, comparable with that encountered in other methods of preservation.

TABLE 46

PROTEIN DIGEST RESIDUE (PDR) TEST OF NET PROTEIN UTILIZATION OF FOODS SUBJECTED TO THERMAL AND RADIATION STERILIZATION[1]

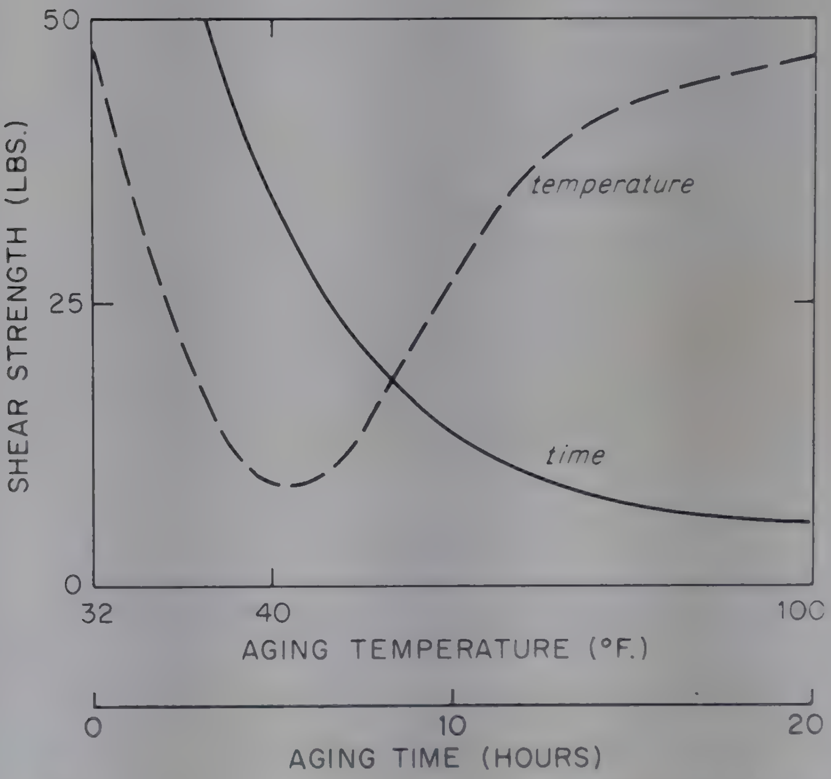
	PDR Index[2]		
Protein	Unprocessed	Thermal Processed	Irradiated[3]
Beef	86	85	87
Pork	88	80	89
Turkey	74	71	74
Evap. milk	75	75	74

[1] From Sheffner, Adachi, and Spector (1957).
[2] Standard errors; beef 0.76, pork 1.29, turkey 0.92, and milk 0.83.
[3] Irradiated at 2 × 10^6 rad level.



Courtesy of and adapted from Drake and Ryer

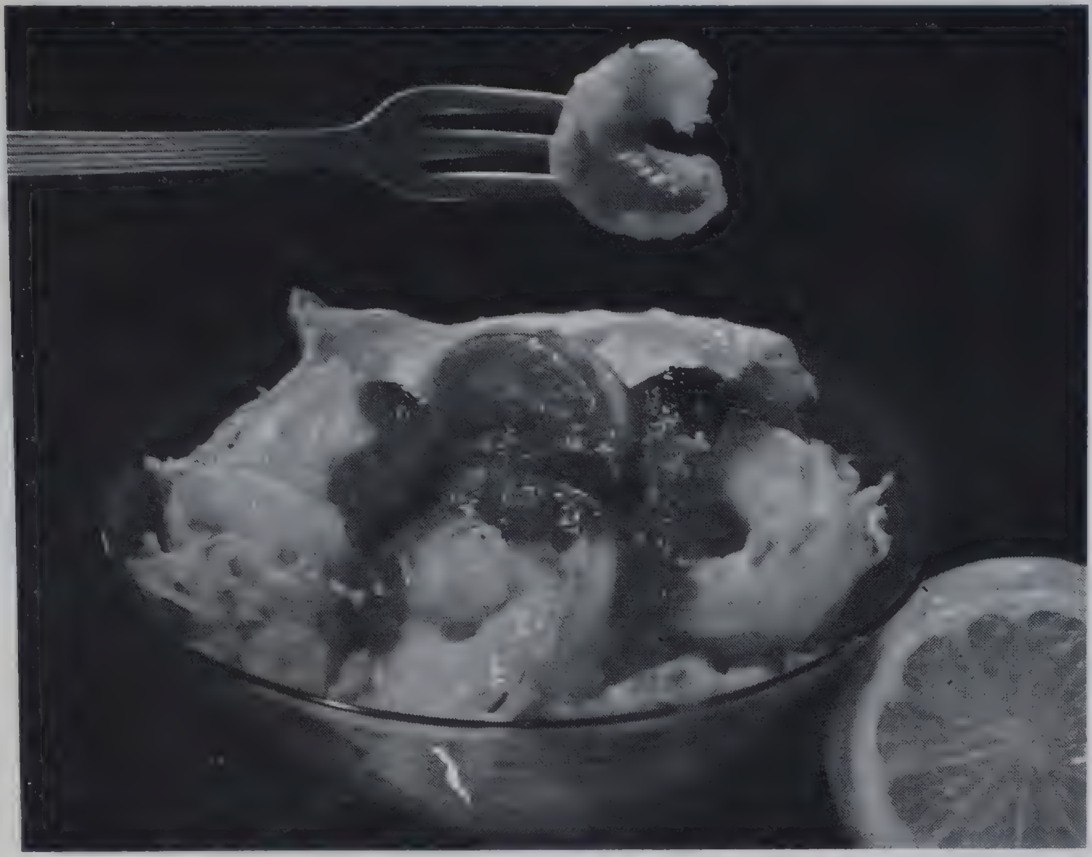
FIG. 94. DECREASE IN RADIATION FLAVOR IN BEEF DURING STORAGE—TEMPERATURE CONDITIONING



Courtesy of Lineweaver

FIG. 95. TENDERNESS CONTROL IN POULTRY MEAT

—induced radioactivity can occur, especially if neutron levels are uncontrolled, or if high energy electrons are employed. Only gamma irradiation sources were used in this experimental program for food preservation, yielding foods with no significant amounts of induced radioactivity. Variations in natural background radioactivity in foods due to geographic areas in which grown, are much greater than any induction which occurs, with due care in radiation source design. It should be mentioned again for emphasis that all living entities on earth are radioactive, so the statements which appear in the literature relative to "absolutely no radioactivity" in foods, irradiated or not, are inaccurate.



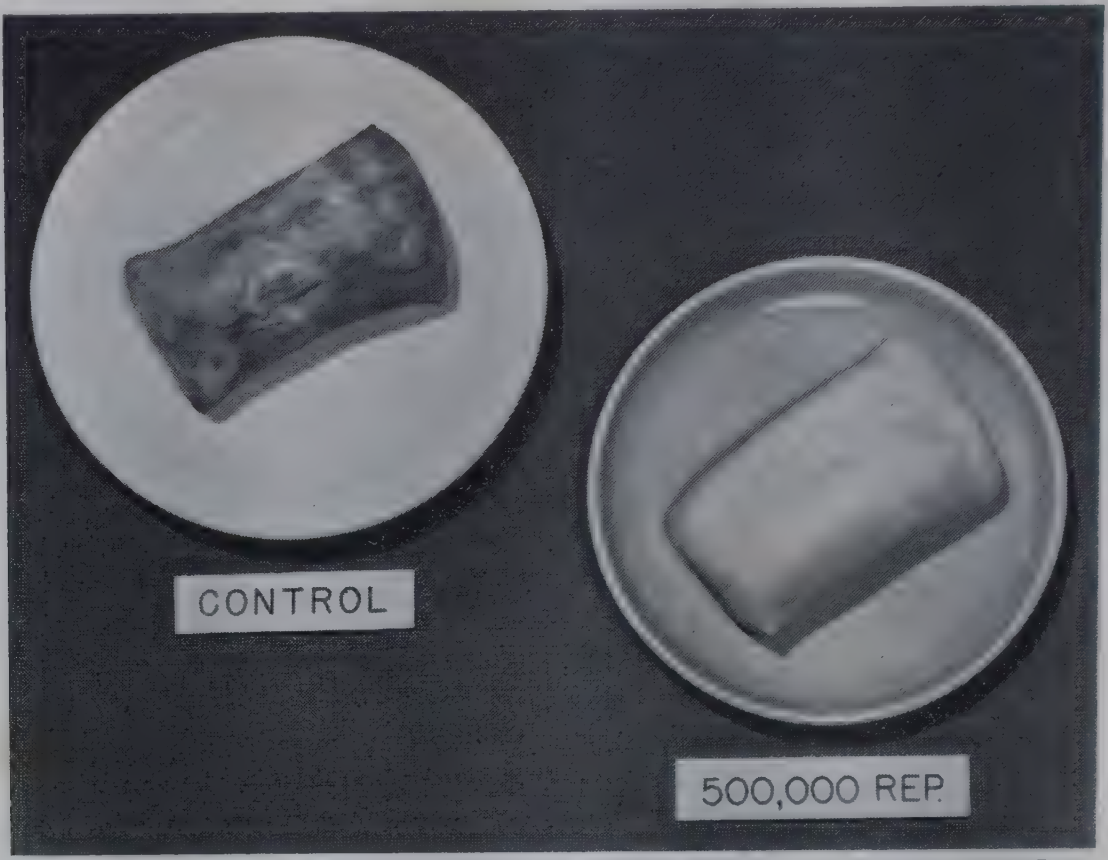
Courtesy of U.S. Army Quartermaster Corps

FIG. 96. RADIATION STERILIZED SHRIMP STORED ONE YEAR AT ROOM TEMPERATURE (4.8 MEGARADS)

From an overall view to date there has been no untoward physiological reaction due to the consumption of radiation stabilized foods uncovered from these studies. There is destruction of nutrients at levels of the same order of magnitude as the destruction by other methods of food preservation.

Considering the youthfulness of the art of radiation processing of foodstuffs, it is to be anticipated that in the near future scientists will uncover methods of protecting nutrients during irradiation much in the same manner as was done in the canning and dehydration and freezing industries.

In summary, the important food classes, including meats, fish, fruits, vegetables, and poultry have been subject to an evaluation and testing program since 1954. Some irradiated foods are reported being distributed in Europe currently.



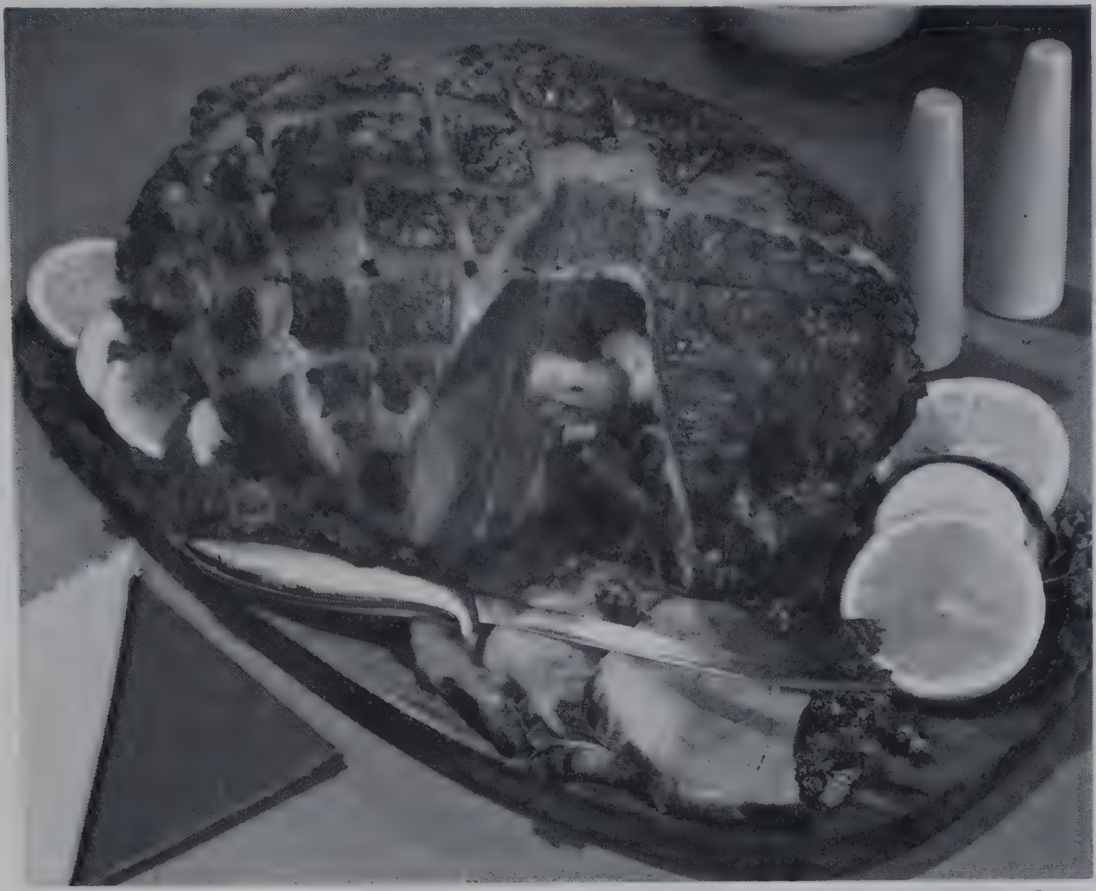
Courtesy of U.S. Army Quartermaster Corps

Fig. 97. Mold Destruction in Bread Rolls

The health and welfare of mankind are intimately interwoven in the fabric called the economics of food distribution. The peaceful applications of atomic energy in the disinfection and stabilization of man's food directly involves the economics of food distribution. Radiation sterilization and radiation pasteurization of foods have direct contributions to make to the health and welfare of all men. The solution to the successful applications of atomic energy in preserving foods requires the best talents men can muster.

Acceptance Studies

The treatment of foodstuffs with ionizing radiations can cause drastic changes in the tastefulness of foods. Providing the technology man has accumulated in food preservation is applied, radiation-stabilized foods can be acceptable. Demonstration of this fact can be seen in the tests conducted by volunteer troops in the U. S. Army. Pork loin, enzyme inactivated with mild heat, given 4.8 million rads of ionizing radiations, in suitable packages, and stored for a year at room temperature was rated equal in acceptance to fresh pork loin by many consumers. Similar results were obtained with chicken. A number of consumers have eaten sterilized bacon, stored a year and more at room temperature, and these consumers were unable to distinguish which item in a meal had been irradiated. While there have been serious off-flavors in radiation treated foods, there is no reason to expect that the mere placing of a food in front of an irradiation source will yield unacceptable products. The addition of odor scavengers, such as a packet of carbon, in containers of food for irradiation, results in greatly improved radiation-stabilized products, after storage. The scavengers are particularly effective with precooked, ready-to-eat foods.



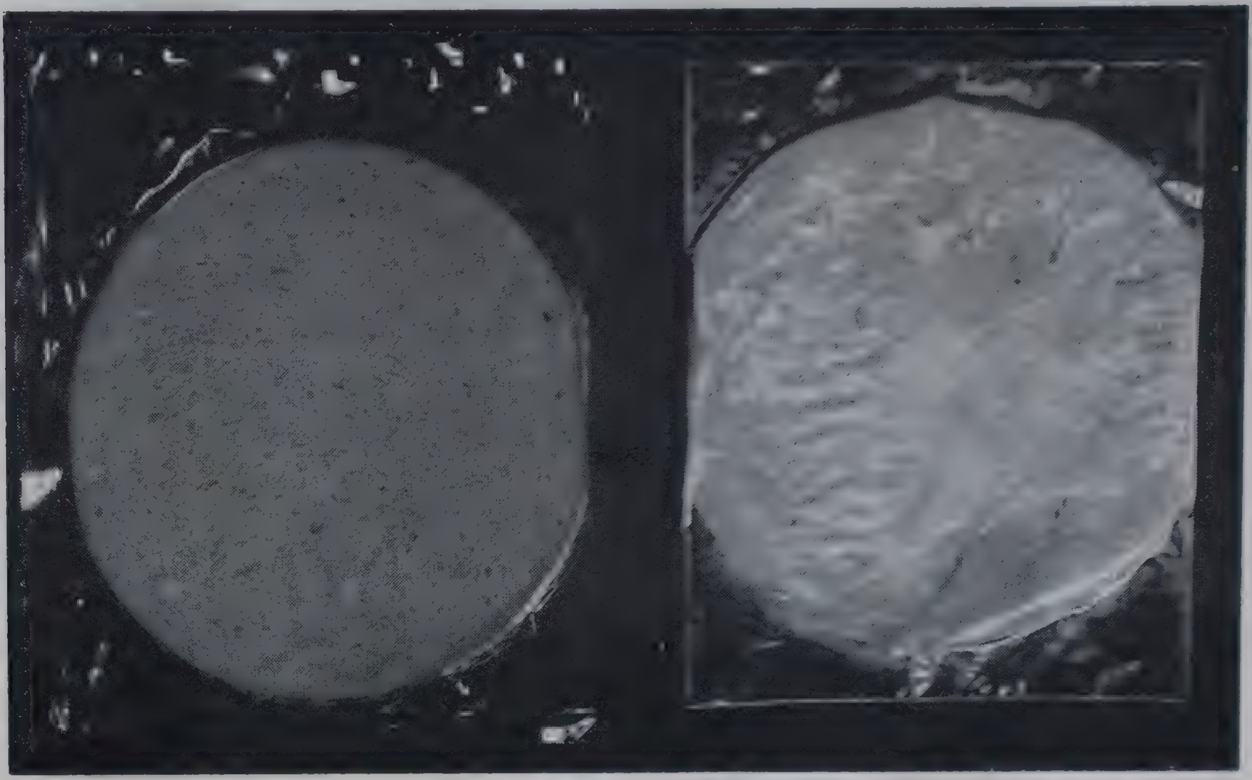
Courtesy of U.S. Army Quartermaster Corps

Fig. 98. Radiation Permits Preservation of Whole Animal or Large Portions of Animals (Ham Shown)

This ham irradiated with 4.8 megarads, stored about eight months at room temperature then cooked.

The off-flavor frequently found in irradiated foods decreases on storage (see Fig. 94). Thus temperature conditioning is most useful and should be an integral part of a successful sterilizing process.

The tenderness of meat given a sterilizing treatment with ionizing radiations is improved. In fact, meat is prone to become too tender. Obvious metabolic methods are available for controlling tenderness of meat in addition to the use of less tender sections of animals, and animals of different grades. The process of rigor mortis can be used in controlling toughness of meats.



Courtesy of U.S. Army Quartermaster Corps

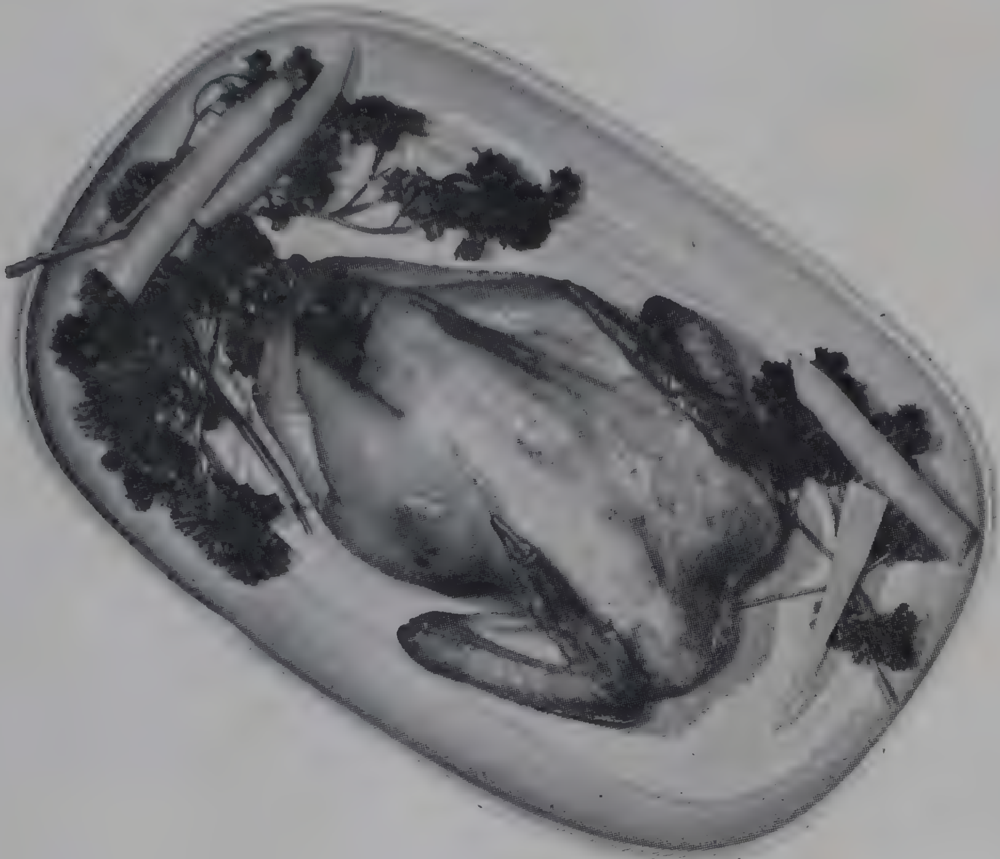
Fig. 99. Improved Refrigerator Storage and Distribution Life of Sliced Meat (After Two Weeks)

Left—irradiated with one megarad; *Right*—Control.
Both stored at 32°F. for several months.

The interference with normal rigor mortis in killed flesh by heat inactivation of enzymes is possible. With poultry, the inactivation of enzymes immediately after killing results in flesh of too rubbery a texture. (See Fig. 95). As a part of the process design for chickens, it is necessary to encourage the subsidence of rigor mortis prior to further treatment for enzyme inactivation and radiation sterilization. In beef, the opposite appears to be desirable. By allowing a degree of aging to occur, it is possible to control the tenderness of beef to some extent.

The texture of fruits and vegetables can be controlled with firming agents in much the same manner as is done in canning and other methods of food preservation.

The juiciness of meat can be controlled. The water binding capacity of the flesh may be improved by application of technologies developed by the meat industry in the curing methods of meat preservation, i.e., the use of phosphates.



Courtesy of U.S. Army Quartermaster Corps

FIG. 100. RADIATION PERMITS ROOM TEMPERATURE STORAGE OF CHICKENS

This chicken irradiated with 4.8 megarads, stored for one year at room temperature then cooked.

Radiation-stabilized chicken has been stored successfully at room temperature for three years to date. Sprout inhibited potatoes have been cold stored in one of the authors' laboratories for two years with excellent results, and storage for three years has been completed to date, although deterioration is marked after three years.

Examples of useful radiation-treated foods are shown in the following photographs. Fig. 96 shows shrimp after a year's storage at room temperature. Fig. 97 shows bread rolls whose storage life has been usefully prolonged. Figs. 98 and 99 indicate the possibilities of preserving whole, large, or sliced portions of animal flesh, ham and bologna being shown. Fig. 100 is of young chicken given a medium-level radiation treatment, allowing the storage for a period at room temperature, although it is not permanently preserved.

It is to be recalled that most of the people on earth have no refrigeration facilities available in homes. In Europe, less than five per cent of the families have a refrigerator.

BIBLIOGRAPHY

Danald, G. E. 1958. What makes radiation "tick"? U. S. Army QM. Food and Container Institute, Activities Rept. *10*, No. 1, 16–20.

Desrosier, N. W. 1959. The Technology of Food Preservation. Avi Publ. Co., Westport, Conn.

Goresline, H. E., and Desrosier, N. W. 1959. Preservation of foods by irradiation. Am. J. Public Health *49*, 488–492.

Jacobs, M. B. 1951. The Chemistry and Technology of Food and Food Products. Interscience Publ., Inc., New York.

Jensen, L. B. 1953. Man's Food. Garrard Press, Champaign, Ill.

Kraybill, H. F. 1955. Nutritive effects on foods sterilized with ionizing radiations. Nutrition Rev. *13*, 193–195.

Kraybill, H. F. 1960. Are irradiated foods harmful? Nucleonics *18*, No. 1, 112–117.

Little, A. D. and Co. 1950. Food Acceptance and Flavor Research. Reinhold Publ. Co., New York.

McNamara, A. T., Conquest, V., Cooney, J. P., Calloway, C. G., and McLaughlin, V. J. 1958. Leaders appraise research in food irradiation. U. S. Army QM. Food and Container Institute, Activities Rept. *10*, No. 2, 57–68.

Mitchell, H. H., and Block, R. J. 1946. Some relationships between the amino acid content of proteins and their nutritive values. J. Biol. Chem. *163*, 599–620.

Morgan, B. H. *et al.* 1957. A status report on the acceptability of irradiated beef. Radiation Project Rept. U. S. Army QM. Food and Container Institute, Chicago, Ill.

Peterman J. D., Tressler, D. K., Morgan, B. H., and Huber, T. E. 1955. The irradiation of foods. U. S. Army QM. Food and Container Institute, Activities Rept. *7*, No. 2, 103–115.

Pilgrim, F. J., and Peryam, D. R. 1958. Sensory testing methods. U. S. Army QM. Food and Container Institute, Chicago, Ill.

Sebrell, W. H., Jr. 1953. Chemistry and the life span. Chem. Eng. News *27*, 3624–3629.

Sherman, H. C. 1946. Chemistry of Food and Nutrition, Macmillan Co., New York.

Tisher, R. G., and Kurtz, G. W 1957. Mechanism of action of ionizing radiations on living matter. U. S. Army QM. Food and Container Institute, Activities Rept. 9, No. 1, 24–27.

U. S. Army QM. Corps 1957. Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

U. S. Army QM. Food and Container Institute 1957. Food preservation by irradiation. Digest of conference at Gatlinburg, Tenn. Jan. 21–25, 1957. Activities Rept. 9, No. 2, 92–102.

CHAPTER 17

Radiation Effects on Packaging Materials

While radiation is capable of killing micro-organisms, parasites, and insects, with the cessation of irradiation nothing prevents the now sterile product from becoming reinoculated. It is therefore important to protect the food or other material from reinfection, and this is accomplished with suitable packaging.

Containers made from plastics, metals, paper, wood, and textiles gradually lose their useful properties through oxidation and other deteriorations when exposed to heat, light, air, moisture, and corrosive gases. It is to be anticipated that ionizing radiation will also have effects on such materials. The deteriorations are dependent on the nature of the materials and the severity of treatment. During the search for materials for constructing nuclear reactors, numerous investigations were undertaken on the effects of high energy radiations on such components. With a background of this information and with the information being uncovered currently in many laboratories, the effects of ionizing radiations on packaging materials are becoming understood.

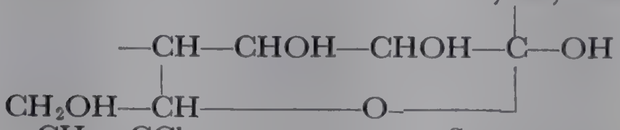
Radiation Effects on Flexible Films

When a polymeric substance is placed in intense radiation field, the polymer may undergo cross-linking or chain scission, both of which alter the characteristics of the material.

In addition, unsaturated bonds are formed, and hydrogen is usually produced, along with traces of other gases, depending on the nature of the polymer. Oxidation will also occur in the presence of air. Several of these reactions may occur simultaneously, the overall effect being governed by the chemical nature of the particular polymer. Such changes have marked influences on the characteristics of plastic films. With cross-linking, there is an increase in molecular weight and a modification of molecular configuration. These result in increased tensile strength, flexural strength, and softening point, and a decrease in elongation. Chain scission, on the other hand, leads generally to decreased tensile and flexural strength. The formation of unsaturated polyene bonds affects the color of the film and, more significantly, increases its susceptibility to oxidation and further chain degradation. Internal gas formation in a plastic increases its porosity. In recent years with the growing knowledge of polymers, the packaging industry has produced packaging materials with most of the qualities desired. The relative stability to ionizing radiation of various polymers of interest are given in Table 47 (Tripp and Crowley 1957).

TABLE 47

RELATIVE STABILITY OF VARIOUS CHEMICAL GROUPINGS[1]

Chemical Grouping	Representative Type Polymer	Dose Required to cause 50% Loss of Tensile Strength 10^9 Rad	Predominant Effect
—$CH(C_6H_5)CH_2$—	Polystyrene	>30	Crosslinking
—$NC_6H_5)CH_2$—	Aniline formaldehyde resins		
—$CH{=}CH$—	Present in elastomers (rubbers)		Crosslinking
—CH_2—CH_2—	Polyethylene	>10	Crosslinking
—CO—NH—	Polyamides such as nylon	>10	
—$Si(CH_3)_2O$—	Methyl silicone		
—$C_6H_3(OH)CH_2$—	Pheno-formaldehyde resin		
—CH_2O—	Polyvinyl butyral	0.6	Crosslinking
—CH_2—S(S)—	Thiokol		Scission
—$C_6H_5C(O)O$—	Polyesters, dacron	4	Crosslinking
—CH_2—CHCl—	Polyvinyl chloride	4	Scission
	Cellulose, sugars, starches		Scission
—CH_2—CCl_2—	Saran	1.0	Scission
—CF_2—CFCl—	Fluorothane		Scission
—CF_2—CF_2—	Teflon	0.07	Scission
—CH_2—CR_2—	Polymethyl methacrylate	0.05	Scission

---

[1] From Tripp and Crowley (1957).

The changes which will occur during irradiation are dependent upon the quantity of energy transmitted to the individual molecule, the rate at which such energy is delivered, and the threshold level of energy required to initiate the various possible reactions. With heat and visible light, the energy imparted by exposures is small, and low threshold level reactions occur, in addition to those possible at all levels requiring less energy, and a range of end products are formed (Bovey 1958).

Polystyrene is markedly resistant to irradiation, and in the absence of oxygen, cross linking occurs, with the evolution of hydrogen gas. The stability of the polymer in a radiation field is explainable in that some 3,000 ev are required to obtain a single cross linkage. This represents a G value of 0.03. Each polystyrene molecule has about 3,000 benzene rings which impart radiation stability to the polymer.

Polyethylene irradiation leads to the disappearance of vinylidene end groups accompanied by the formation of vinylene groups, and evolution of hydrogen gas and short chained unsaturated hydrocarbons. Some cross linking occurs. The effects of irradiation on the functional qualities of polyethylene are shown in Table 48. At the dosages of interest in sterilization, less than six million rads, only insignificant changes in tensile strength, elongation, greaseproofness, moisture vapor transmission rate, oxygen permeability or film sealability have been noted. The main difficulty with polyethylene resides in the gaseous products released during irradiation, which contribute slight off odors and flavors (Tripp 1954). These gaseous products are influenced by the presence of oxygen. In many cases the odors are referred to as paraffin-like, and occasionally as a butyric odor. It has been reported by Tripp (1957) that the age of the polymer, or "freshness" of the packaging film is critical, off odors and flavors being observed mainly in film stored and aged prior to irradiation. Further, some oxidation products are leachable by hot water from such aged film not subjected to irradiation. It is quite probable that irradiation accelerates some of the oxidation which has already started in natural film aging. Further, the usefulness of unirradiated aged film as a packaging material is itself questionable because of these reactions.

TABLE 48

EFFECT OF HIGH ENERGY RADIATION ON THE FUNCTIONAL PROPERTIES OF POLYETHYLENE[1]

Dose megarep	Tensile strength lb./sq. in.	Ultimate elongation per cent	Greaseproofness seconds	Moisture vapor transmission rate[2]	Sealability after irradiation tensile strength of seal lb./in. width	Oxygen permeability cc./100 sq. in./24 hr. sample 0.002 inch thick
0	2100	650	>1800	1.24	3.98	317
1.0	2180	635		0.99	3.56	
3.0			>1800	1.24	4.83	340
6.0				1.13	3.41	
10.0	1729	500				

[1] From Tripp and Crowley (1957).

[2] 100°F. 90% R.H. in g./100 sq. in./24 hr./0.001 in. thickness.

Mylar,[1] known chemically as polyethylene terephthalate, is quite stable in a radiation field; while cross linking does not usually occur, some scission is found. In the absence of oxygen, increasing carbonyl groups are formed. Because of the difficulty of sealing Mylar, it has been used in conjunction with other films as laminates and coatings which yield acceptable sealing characteristics. Polyethylene coated Mylar has been found functional, and in radiation fields the minor effects on the polyethylene component are so low that the package is acceptable for many irradiated foods. The effects of irradiation on such a composite film are shown in Table 49. In the presence of fatty foods, seals are weakened

---

[1] Mylar is a registered trade mark of E. I. du Pont de Nemours and Co.

TABLE 49

PACKAGE PERFORMANCE CHARACTERISTICS

Radiation Dose = 6 million rads; + = satisfactory; − = unsatisfactory; ? = requires determination

Performance Criteria		Metal Cans Enamel-Lined	Videne	Poly-styrene	Mylar	Nylon	Scotchpak (poly-ethylene-coated polyester)	Polyvinyl wax chloride (dip coating)	
Moisture vapor transmission rate 0.002 g/100 sq. in./24 hrs./100 °F.—90 per cent R.H.		+	+[1]	+[1]	+[1]	+[1]	+[1]	+[1]	?
Oxygen transmission rate 0.002 cc./100 sq. in./24 hrs.		+	+[1]	+[1]	+[1]	+[1]	+[1]	+[1]	?
Change in physical properties (−10 per cent)		+	+	+	+	+	+	+	+
Change in chemical properties (precise limits not determined)	odor	+	+	+	−	+	−	−	+
	flavor	+	+	−	+	?	−	−	?
	extractives	?	?	?	?	?	−	−	?
7-foot drum[4] 225 falls	ambient conditions	+[2] ?[3]	?	?	+	?	+	?	?
	−40 °F.	+[2] ?[3]	?	?	+	?	+	?	?
6–3-foot face drops[4]	ambient conditions	+[2] ?[3]	?	?	+	?	+	?	?
	−40 °F.	+[2] ?[3]	?	?	+	?	+	?	?
Sealability		+	?	?	−	−	+	+	+
Stress crack	1,000 hrs.	+	?	?	?	?	+	+	?

[1] Based on foil laminations. [2] Tin-plated steel. [3] Aluminum. [4] Packed shipping containers weighing 50 lbs.

* Adapted from Tripp (1958)

and some delamination is found, the degree being directly related to the dose applied and the temperature of storage. Polyethylene coated Cellophanes behave in a similar manner.

The irradiation of Pliofilm at sterilizing dose levels (five megarads) does not alter moisture-vapor-transmission rates, but losses occur in tensile strength.

Halogenated plastics including Saran, teflon, polyvinyl chloride and fluorothene have a major drawback because of release of halogen gases and halogen acids on irradiation. The rate of evolution is related to the dose applied.

Variable reports have been made about Saran films, perhaps the age of the film, its formulation, and previous history influencing the results obtained following irradiation. The formation of halogen acids promotes degradation of the package during storage and these are objectionable. The effects of irradiation of Saran at five megarads results in no change in moisture vapor transmission rate but a decrease of 20 per cent is found in tearing resistance.

Cellulose and cellulosic materials occupy a prominent position among packaging materials including papers which are waxed, coated, or laminated. Scission of the cellulose molecule occurs during irradiation. At very high doses cellulose can be made water soluble (Table 50). Degradation of cellulose decreases the tensile strength and tear resistance of papers and Cellophanes. (Table 51, 52).

TABLE 50

DECOMPOSITION OF CELLULOSES BY IRRADIATION[1]

Cellulose material	Dose-megarep	Decomposition of Carbohydrate per cent	Over-all sugar yield per cent
Cotton linters	0		23.0
	10	2	30.4
Wood pulp	0		33.5
	10	5	44.5
Wood	0		25.0
	10	3	36.5
Glucose	10	2	

[1] From Tripp and Crowley (1957).

TABLE 51

TENSILE STRENGTH OF IRRADIATED CELLULOSIC MATERIAL (POUNDS/INCH OF WIDTH)[1]

Dose megarep	35 pound kraft coated with 18 pounds polyethylene	450 gauge MSAT cellophane	MSB-3 cellophane	Bleached glassine
0	48.38	24.4	24.8	13.4
2	44.65			
3.5		16.0	21.8	14.5
5	40.86			
10	39.61			

[1] From Tripp and Crowley (1957).

The properties of laminants dependent upon cellulose fiber strength, such as strength of seal and peeling are also affected (Table 53). Greaseproofness, on the other hand is not seriously reduced by irradiation. The

TABLE 52

INTERNAL TEARING RESISTANCE OF IRRADIATED CELLULOSIC MATERIALS (GRAMS)[1]

Dose megarep	35 pound kraft coated with 18 pounds polyethylene	300 gauge MSAT-83 cellophane	450 gauge MSAT-80 cellophane	300 gauge MSAT-54 cellophane
0	93	11	14	17
3.0	84	8	6	6

[1] From Tripp and Crowley (1957).

effect of the material being packaged on the functional characteristics of irradiated films can be seen in Table 55 for cellophane.

Paraffin is commonly used in packaging to provide a moisture barrier, a sealing agent, or a laminating agent. Paraffin has a structure similar to polyethylene, differing in molecular weight and degree of branching. Upon irradiation, paraffin reacts much as does polyethylene. Some cross linking occurs, and insoluble gels are formed.

Lamination is commonly used in food packaging materials. Usually a plastic film forms the inside surface in contact with the food, next comes a layer of aluminum foil, then a layer of paper, or another film to form the outer layer. The plastic film provides the heat sealing quality and chemical protection to the aluminum foil; the foil contributes moisture and gas penetration resistance; the outer layer contributes the mechanical strength. Irradiation of such a laminated film has effects on all components, including the adhesive used to bond the layers together. The ad-

TABLE 53

TENSILE STRENGTH (PEEL) OF IRRADIATED SEALS (35 POUNDS PER REAM KRAFT PAPER COATED WITH 18 POUNDS PER REAM POLYETHYLENE)[1]

Dose megarep	Tensile strength, lb./in. width
0	3.71
2	3.36
5	3.26
10	2.81

[1] From Tripp and Crowley (1957).

TABLE 54

EFFECT OF RADIATION ON THE GREASEPROOF OF CELLULOSE MATERIALS (GREASEPROOFNESS IN SECONDS)[1]

Cellulose material	Dose 0 rep	Dose 3 megarep
Waxed paper (26 pound sulfite—9 pound paraffin wax)	340	320
35 pound kraft coated with 18 pounds polyethylene	>1800	>1800
Bleached glassine	>1800	>1800
P-1 Cellophane	>1800	>1800
MSB-3 Cellophane	>1800	>1800

[1] From Tripp and Crowley (1957).

TABLE 55

MOISTURE VAPOR TRANSMISSION RATE OF IRRADIATED CELLOPHANE (G./100 SQ. IN./24 HRS. 100°F.—90 % R.H.)[1]

Grade cellophane	Contents of package	Dose megarep 0	3.5	7.0
MSAT-87	none	1.1	1.4	2.4
300 gauge	water	2.5	3.2	7.8
MSAT-80	none	1.7		3.8
300 gauge	water	2.9	4.5	8.0

[1] From Tripp and Crowley (1957).

TABLE 56

LOSS OF ADHESION OF VINYL COMPONENT DURING IRRADIATION[1]

Dose megarep	0.0015-inch thick vinyl laminated to 0.001-inch aluminum laminated to 44 × 36 scrim peel strength vinyl component lb./in. width
0	0.84
10	0.028

[1] From Tripp and Crowley (1957).

hesives are susceptible to radiation damage, and to substances which may be released by the laminant plies. This is shown in Table 56.

At doses of two million rads and less, radiation has no significant effects on the physical characteristics of flexible containers. At doses over two million rads, changes occur in physical properties of plastic films such as polyethylene, mylar, vinyl, and polystyrene, but the changes are of minor consequence. Saran, Pliofilm, and Cellophanes show embrittlement at doses over 3 million rads. (Table 57–63).

TABLE 57

FAILURES OF FLEXIBLE PACKAGES (PER CENT)[1]

Package	Contents	Dose (Megarad) 0	1	3	6
Polyethylene (H.D.)	Lactic acid sol.	40	—	—	—
	Salad oil	25	—	—	—
Nylon (Caprolactam)	Lactic acid sol.	80	80	83	83
Polyvinyl chloride	Salad oil	20	20	20	20
—Fluorothane	Salad oil	20	20	50	100
Polyethylene laminated to	Lactic acid sol.	0	33	20	—
Cellophane	Salad oil	100	—	—	—
Polyethylene coated Mylar	Lactic acid sol.	25	35	50	35
Saran 6D	Lactic acid	80	95–100	95–100	95–100

[1] From Tripp (1958).

TABLE 58

REVOLVING DRUM TESTS OF FLEXIBLE CONTAINERS PACKED IN SHIPPING CONTAINERS[1]

Package Material		Falls to Failure (leakage) Min.	Max.
Polyethylene coated	Test 1	340	450
Polyester film type S	Test 2	509	600+
Polyester, type V (0.0005 inch) Laminated to aluminum (0.001 inch) Laminated to polyester, Type M (0.0005 inch)		230	270

[1] From Tripp (1958).

TABLE 59

DYE PENETRATION AFTER FLEXING (GELBO TESTER)[1]

Package	Number of Flexes Before Failure
Saran (0.002 inch thick)	300
Polyethylene (low density 0.002 inch thick)	500+
Polyethylene (0.002 inch) coated polyester (0.00005) Type S	200
Polyethylene (0.002 inch) coated polyester (0.001 inch) Type M	500+
Polyvinyl Chloride (0.0015 inch) laminated to aluminum foil (0.001 inch) laminated to polyester (0.0005 inch) Type M	50

From Tripp (1958).

TABLE 60

GASEOUS PRODUCTS (MICROMOLES/GM. SAMPLE) (DOSE 6 MEGARADS)[1]

	Polyethylene (low density)		Polyethylene (high density)		Fluorothane	
	Vacuum	Air	Vacuum	Air	Vacuum	Air
Carbon Dioxide	0.3	0.3	0.1	0.1	27.9	27.7
Oxygen	0.1	0.1	0.1	0.1	0.3	0.4
Nitrogen	1.5	1.5	0.8	0.8	8.5	8.1
Water	0.5	3.8	0.2	1.1	0.5	2.1
Hydrogen	95.7	93.6	98.7	94.6	0.6	—
Silicon tetrafluoride	—	—	—	—	59.9	57.5
Hydrogen chloride	—	—	—	—	2.3	—
Ethylene	1.2	—	—	—	—	—
Acetone	0.3	—	—	3.2	—	4.3
Butene	0.1	0.6	—	—	—	—
Propene	0.3	—	0.1	—	—	—

[1] From Tripp (1958).

TABLE 61

GASEOUS PRODUCTS (MICROMOLES/GM. SAMPLE) (NOT CORRECTED FOR RESIDUAL AIR) (DOSE 6 MEGARAD)[1]

	Saran		Mylar	
	Vacuum	Air	Vacuum	Air
Carbon dioxide	6.2	1.3	0.1	0.5
Argon	—	1.1	0.9	1.0
Oxygen	—	4.8	20.3	18.6
Nitrogen	5.5	92.0	78.4	79.4
Water	5.1	0.4	0.3	0.3
Hydrogen	5.2	0.1	—	0.2
Silicon tetrafluoride	1.1	—	—	—
Acetic acid	12.2	—	—	—
Ethanol	0.6	—	—	—
Hydrogen chloride	43.5	0.3	—	—
Hydrogen cyanide	18.4	—	—	—
Ethylene	2.2	—	—	—

[1] From Tripp (1958).

TABLE 62

ODOR INTENSITY OF IRRADIATED PLASTICS[1]

Plastic	Dose Megarad				
	0	1	3	6	Av.
Polyethylene (no additives)	1.6	3.2	3.6	4.7	3.3
Polyethylene (commercial)	1.4	4.0	5.2	5.6	4.1
Polyethylene (2.5 per cent carbon black plus anti-oxidant)	1.6	4.2	5.5	6.0	4.3
Polyethylene (6.0 per cent carbon black plus anti-oxidant)	2.6	4.4	5.2	5.4	4.4
Polyethylene (H. D. Ziegler process)	1.7	3.8	3.6	3.9	3.2
Wax (Fischer Tropsch)	2.0	3.7	4.6	5.3	3.9
Polystyrene	3.0	2.4	2.8	1.6	2.5
Nylon 6	1.9	1.4	1.7	1.4	1.6
Mylar	2.2	2.0	2.6	2.5	2.3
Videne	1.6	2.5	2.4	2.1	2.2

[1] From Tripp (1958).

TABLE 63

NON-VOLATILE WATER SOLUBLE SUBSTANCES IN IRRADIATED POLYETHYLENE (H.D.) (MICROGRAMS/GM. OF FILM)[1]

Dose in Megarad			
0	1	3	6
27.4	29.8	31.2	37.1

[1] From Tripp (1958).

Irradiation of most foods in some plastic containers results in off flavors and odors in the food. Nylon is very low in odor formation during sterilizing irradiation. Polyethylene irradiation at sterilizing doses evolves slightly objectionable odorous compounds (short fragments of polymer), which are carried into the food itself. Off odors may be removed in the package with odor scavengers.

Thin plastic films are objectionable due to the ability of micro-organisms to penetrate through microscopic openings, caused by damage during sealing and rough handling. This may be overcome by increasing the thickness and improving sealing techniques.

Laminated metal foils and plastics, and combinations of the latter, are functional (Tripp 1958). Some containers, such as Scotchpak, are as durable in rough handling as metal cans, and perform satisfactorily up to six megarads as a food package. This performance of standard-type container materials is given in Table 49.

Radiation Effects on Rigid Materials

The commercially available tin plated steel cans used in canning and freezing are the most successful package currently for irradiated foods and have been the most widely used container.

Rigid metal containers such as tin and aluminum cans have been highly developed. Tin coated containers have been used successfully for more than a century for sterile foods. The aluminum containers have become more widely used in Europe than in the U. S. A. and such containers are continuing to be perfected.

At sterilizing dose levels, steel is stable. At doses ranging from 60,000,000 rads and higher, damage occurs in steel. The effect on aluminum is similar. Radiation has no influence in promoting tin rot or tin disease which is the transition of rhombic to cubic crystalline structure of tin. Trace amounts of bismuth prevent this transition, in any event. Tin coatings over base steel are suitable for food irradiation. End sealing compounds generally used in metal containers are actually improved slightly by irradiation. The exceptions are found in butyl rubber sealing compounds which are apparently depolymerized by irradiation. Of the interior can enamels for tin plated cans, the oleoresinous enamels are unsatisfactory for high fat foods. Oleoresinous enamels appear satisfactory for enzyme inactivated foods. Bonding of enamels to rigid aluminum containers causes some difficulty. Except for this, similar enamels may be used for tin and aluminum containers (Tripp 1958).

The metal containers have been found to be the most satisfactory of the packaging materials available to date. Irradiated foods stored for three years at room temperature have been found highly acceptable when tin plated steel cans were used. In general, there are no significant differences in the cans containing heat treated or irradiated foods after a year or two of storage at room temperature.

Glass containers have not received much attention in research as irradiated food packages due perhaps not only to their fragility, but also to the discoloration which occurs in irradiation. The discoloration is reported to be minimized by the addition of cerium.

Container Shape

Container shape is important. Ideally cubic forms would be desirable for best radiation source utilization and dose distribution and control. But cylindrical rigid containers have found widespread use in the canning industry. Cylindrical cans are therefore the most commonly available, although rectangular cans are used for certain meat items such as luncheon meat loafs and sardines.

Other Packaging Considerations

Radiation processing of foods permits the storage of otherwise perishable commodities of high moisture content in metal and plastic containers at room temperatures. Flexible containers have been developed for frozen and refrigerated foods, the latter of short duration in storage. Functional, economical plastic containers are being developed and should find application in radiation sterilization and pasteurization.

The shape of the container used in the radiation process is dictated somewhat by the configuration and organization of the source, e.g., high energy electrons have limitations in depth of penetration. It is seen that the overall dimensions of the container to be irradiated are determined by the attenuation of the radiation in the container and contents, and the maximum dose variation that can be tolerated by both. Dose distribution and efficiency are affected by container construction materials. Tripp and Crowley (1957) have listed radiation absorber densities for container materials, shown in Table 64.

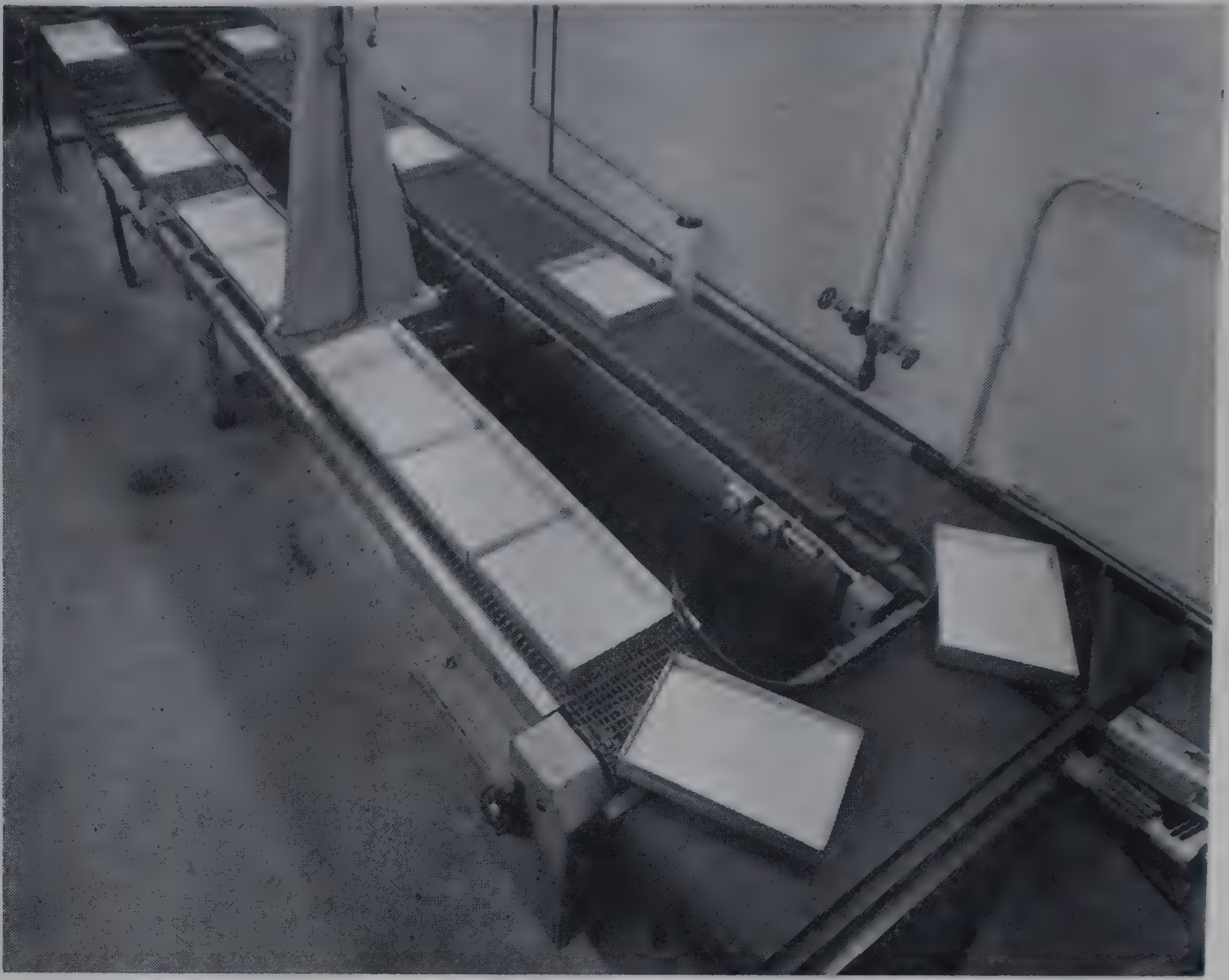
TABLE 64

ABSORBER DENSITIES OF SOME PACKAGE MATERIALS[1]

Package		Absorber Density g./sq. cm.
Papers:		
18-lb. per ream kraft or sulfite		0.003
26-lb. per ream kraft or sulfite		0.0035
26-lb. per ream kraft or sulfite coated with 9-lb. per ream paraffin		0.005
40-lb. per ream kraft or sulfite		0.006
60-lb. per ream kraft or sulfite		0.009
Bending Boards:		
33-lb. per 1,000 sq. ft.		0.016
65-lb. per 1,000 sq. ft.		0.032
90-lb. per 1,000 sq. ft.		0.044
Plastic Films:		
Polyethylene	0.001-inch thick	0.0023
Saran	0.001-inch thick	0.0044
Polyvinyl chloride	0.001-inch thick	0.0035
Pliofilm	0.001-inch thick	0.0028
Mylar	0.001-inch thick	0.0035
Cellophane	300-gauge MST-53	0.0035
Cellophane	450-gauge MSAT-80	0.0049
Metals:		
Aluminum	0.012-inch thick	0.0824
Steel	60-gauge (0.0066-in. thick)	0.134

[1] From Tripp and Crowley (1957).

The failures found in flexible packages containing food systems are shown in Table 57. The ability of flexible containers to withstand rough handling is indicated in Table 58. Flexing alters the characteristics of flexible films, and by using dyes it is possible to estimate changes in integrity of films (Table 59). The irradiation of films leads to gaseous products, and those identified following the irradiations of selected films are shown in Table 60 and Table 61. The volatiles which result from an irradiation treatment vary from film to film. With increasing dose it is found that relatively more intense odors are detectable (Table 62). The non-volatile



Courtesy of High Voltage Engineering Corp.

FIG. 101. ELECTRON STERILIZATION OF PLASTIC CONTAINERS

water solubles which are present and which may leach from the film to the food or material being irradiated are indicated in Table 63.

The successful application of radiation in the sterilization of large packaging containers is found commercially today (Fig. 101).

BIBLIOGRAPHY

BALLANTINE, D. S., DIENES, G. J., MANOWITZ, B., ANDER, P., and MESROBIAN, R. B. 1954. Evaluation of polyethylene cross linked by ionizing radiation. J. Polymer Sci. *13*, 410–414.

BOPP, C. D., and SISMAN, O. 1955. Radiation stability of plastics and elastomers. Nucleonics *13*, No. 7, 28–33.

BOPP, C. D., and SISMAN, O. 1956. Stress strain values for reactor irradiated plastics. Nucleonics *14*, No. 3, 52–59.

BOVEY, F. A. 1958. The Effects of Ionizing Radiation on Packaging Materials and Synthetic High Polymers. Interscience Publ. Co., N. Y.

BRATZLER, L. J. 1958. Dip coat packaging of irradiated foods. QM. Food and Container Institute. Research Report S-584, Chicago, Ill.

COLLINS, H. E. 1958. Design of aluminum containers for irradiated foods. QM. Food and Container Institute. Research Report S-734, Chicago, Ill.

DESROSIER, N. W. 1959. The Technology of Food Preservation. Avi Publ. Co., Westport, Conn.

FEAZEL, C. E. 1958. Determining the functional properties of food containers used for foods irradiated in combination with model food systems. QM. Food and Container Institute Research Report S-554, Chicago, Ill.

HATHAWAY, C. T. 1958. Evaluation of films for irradiation of packaging of foods. QM. Food and Container Institute. Research Report 3–744, Chicago, Ill.

LARSON, W. S. 1958. Materials for containers for irradiated food packages. QM. Food and Container Institute. Research Report S-556, Chicago, Ill.

LITTLE, K. 1952. Irradiation of linear high polymers. Nature *170*, 1075–1079.

O'BRIEN, R. 1958. Development of aluminum containers for irradiated foods. QM. Food and Container Institute. Research Report S-733, Chicago, Ill.

PRATT, G. B., and WILBUR, A. C. 1958. Corrosion Studies of irradiated canned foods. QM. Food and Container Institute. Research Report S-700, Chicago, Ill.

PRATT, G. B., and WILBUR, A. C. 1958. Effect of radiation on can enamels. QM. Food and Container Institute. Research Report S-567, Chicago, Ill.

PROCTOR, B. E., LOCKHART, E. E., and GOLDBLITH, S. A. 1954. The application of electronic treatments to the destruction of insects in packaged military rations and packaging materials. Final Report, U. S. Army QM. Corps Contract DA 11-009-QM 1988, Chicago, Ill.

SMITH, J. E. 1958. Investigation of the effects of radiation on waxes and wax coated paper products. QM. Food and Container Institute Research Report S-748, Chicago, Ill.

SUN, K. H. 1954. Effects of atomic radiation on high polymers. Modern Plastics 32, 141–151.

TRIPP, G. E. 1954. Effect of cathode rays on subsistence packages made of polyethylene coated kraft paper. Progress Report, U. S. Army QM. Corps Project 7-84-01-002.

TRIPP, G. E. 1957. Packaging of irradiated foods. Chap. 28. Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

TRIPP, G. E., and CROWLEY, J. P. 1957. Effect of radiation energy on flexible containers. Activities Report, 9, No. 2, 112–122. Quartermaster Food and Container Institute, Chicago, Ill.

TRIPP, G. E. 1958. Packaging for irradiated foods, U. N. F. A. O. International Meeting on Radiation Preservation of Foods. Harwell, England, Nov. 11, 1958.

UNITED STATES ARMY QM. CORPS. 1957. Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

Food Irradiation Technology

While individual studies on the application of radiation to the destruction of micro-organisms, parasites, and insects were found in the literature early in the 20th Century, it was not until the middle 1950's that large scaled efforts were directed to exploiting radiation as a sterilizing agent.[1] Work by Proctor and associates at the Massachusetts Institute of Technology, Huber and associates in New York, and others in the late 1940's laid the groundwork which indicated the technical feasibility of this use of radiation. Based on preliminary studies, the U.S. Atomic Energy Commission supported a series of investigations in the early 1950's with the primary goal of establishing industrial radiation sterilization. An intensive program designed to bring about the early application of this peaceful use of atomic energy soon followed, not only in the U.S.A., but in several other countries as well.

In the U.S.A., the QM. Food and Container Institute for the Armed Forces in Chicago, Illinois, was nominated as the governmental agency responsible for the research and development program in food irradiation, and it was assisted by the Atomic Energy Commission. The Surgeon General of the U.S. Army was responsible for the wholesomeness studies with irradiated foods; these studies were coordinated with the Food and Drug Administration of the Department of Health, Education, and Welfare. All tests were so coordinated that when the research results are evaluated, simultaneous clearance and approval of irradiated foods is anticipated for both military and civilian personnel in the U.S.A. Early leadership was under Siu, Tischer, Morgan, and Pomerantz of the U.S. Army QM. Corps, and Huber of the Office of the Surgeon General of the U.S. Army.

In the U.S.A. more than 300 scientists in more than 120 laboratories participated in this research coordinated with the aid of the National Academy of Science—National Research Council.

About 80 European scientists in 13 laboratories independently are conducting research in this area, mainly on low level irradiation of foods to improve distribution and marketing practices. U.S.S.R. scientists in smaller numbers are also active in this field.

The results to date from food irradiation research can be summarized as follows:

---

[1] Sale of irradiated food is not yet permitted (June, 1960) by the U.S. Food, Drug, and Cosmetic Administration.

1. Many foods can be successfully stabilized with radiation. With the present rate of expanding technology, most foods should soon fall within the successful category.
2. Suitable packages are available for irradiated foods. Functional plastic films are becoming available.
3. The nuclear physics problems relating to the process have been or are being resolved.
4. Radiation stabilized foods are being established as wholesome and useful for mankind to consume.
5. Consumers are generally unable to tell which item in a meal is irradiated provided the best available techniques of irradiation are used.
6. There are many other areas of application which could profitably be investigated.

While radiation sterilized foods have important uses in national defense and in natural disaster circumstances, major industrial interest in the U.S.A. is found in low level irradiations, the so-called pasteurization ranges of less than a million rads for consumer items (Clifcorn 1955, McNamara *et al.* 1958). Extension of useful refrigerated storage and distribution life of perishable foods by factors of five to ten compared with conventional methods may be obtained (Brownell *et al.* 1955). Major attention has been given to the 22 foods listed in Table 42, page 288, examples to be given below.

The successful high level radiation stabilization treatment of foods requires the following:

1. Stabilization of biochemical systems by inactivating enzymes with heat and controlling oxidation. The stabilization problems are product specific.
2. Packaging the foodstuffs to prevent recontamination. The packaging requirements are product-specific.
3. Administration of sterilizing doses of ionizing radiations with suitable radiation sources, to control micro-organisms and minimize induced radioactivity. The radiation requirement for sterilization is product-specific.

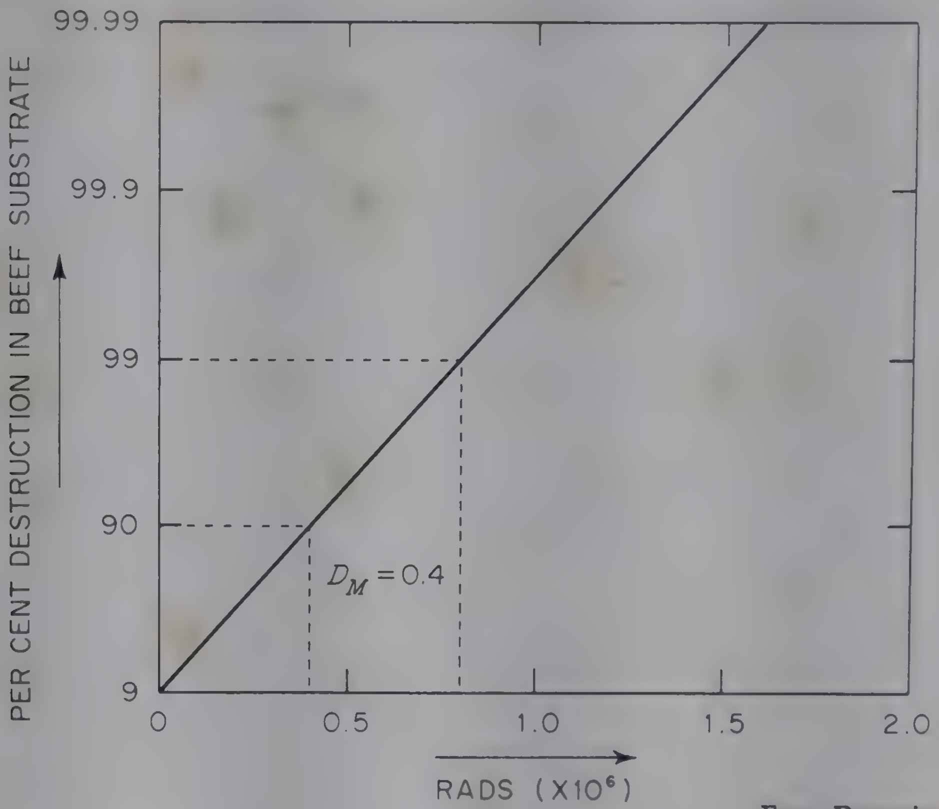
General Method for Establishing a Radiation Stabilization Process for Foods

Food may be stabilized by inactivating the micro-organisms and enzymes present, and protecting the stabilized food from recontamination and access to oxygen. The latter problem areas are controlled by suitable packaging. The irradiation of a food can destroy micro-organisms and enzymes. It is far more efficient to employ ionizing radiations to kill micro-organisms (Fig. 102) than to inactivate enzymes (Fig. 103). When inactivation of enzymes is necessary, it is desirable to inactivate them by other means, to complement the irradiation action.

The sterilization of foods with ionizing radiations involves two major considerations—the food product and a suitable radiation source.

The Food Product—Micro-organism Destruction

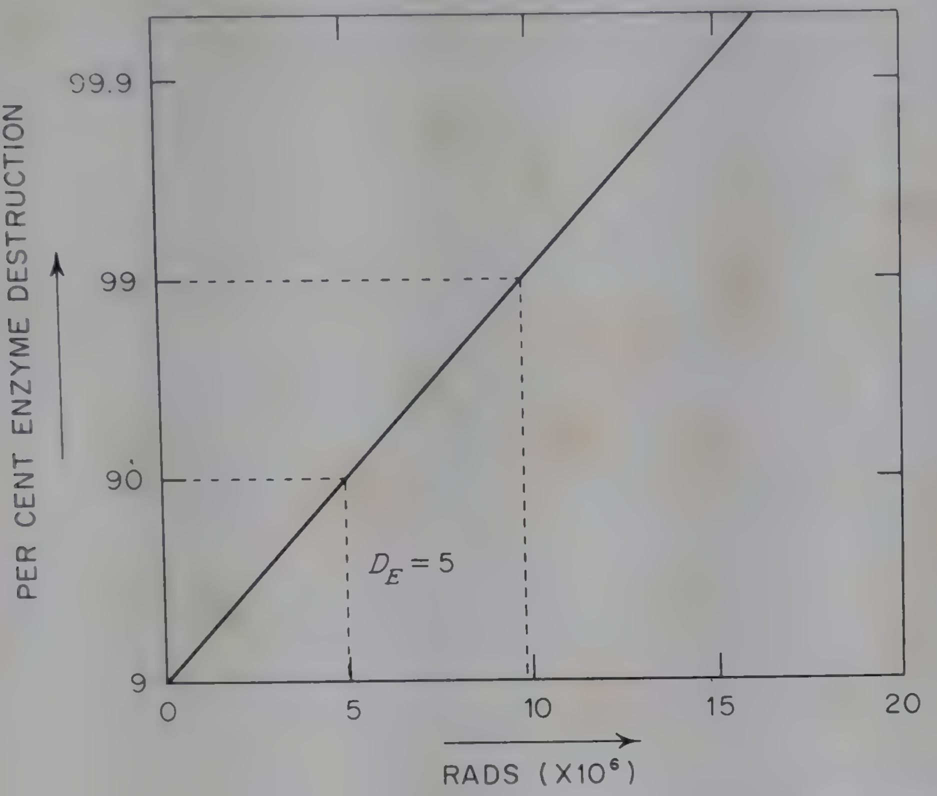
The characteristics of the food itself dictate the types of spoilage organisms which are capable of spoiling the food. As noted previously, the foods are classed as acid (pH values below 4.5) and low acid (pH values above 4.5). Above pH 4.5, *Cl. botulinum* must be assumed present, as is done in designing the canning process. *Cl. botulinum* is the most radiation resistant spoilage organism causing difficulty. In canning, putrefactive anaerobes and certain thermophilic organisms are more heat resistant than *Cl. botulinum.* Due to its radiation resistance, the sterilization dose requirement for foods with pH values greater than 4.5 is that dose necessary to reduce the probability of having live spores of this organism surviving the designed process. The degree of safety required has been found to be in the order of one chance in a billion of a spore surviving. In practice, it has been found that spore destruction with ionizing radiations follows a logarithmic order of death.



From Desrosier

FIG. 102. THE D_M VALUE FOR *Clostridium botulinum*

It is useful if the "D_M" value is defined as that dose giving a 90 per cent reduction in microbial populations (see Fig. 102). If we assume that the food contains originally 1,000 spores per gram and that there may be 1,000 grams in each can, then it follows that, if 12 D_M values are accumulated, there will be only one chance in a billion that any can will contain live spores. There are obviously safety margins involved, first in the inoculum of spores used in establishing the D_M value, second, the low level of actual contamination by *Cl. botulinum* in food, and third, the general sterility of internal tissues of a food (usually the contamination, if it occurs, is on the exposed surfaces of foods).



From Desrosier

FIG. 103. THE D_E VALUE FOR ENZYMES

The D_M value for *Cl. botulinum* has been established to be 400,000 rads in foods with pH values greater than 4.5 (Goresline and Desrosier 1959). The sterilization dose is therefore 12 × 400,000, or 4.8 million rads. The sterilization power of ionizing radiations can be altered by the temperature of irradiation, the dose rate applied, and through changing the substrate by addition of chemical entities which sensitize spores.

The 12 D_M value for *Cl. botulinum* of 4.8 million rads is the dose necessary to kill the most resistant strains of the organisms. Generally type A strains are more resistant than type B, and type B is more resistant than types C and E.

For foods with pH values below 4.5, the 12 D_M value has been established for the most resistant acid food spoilage organism, be it a bacterium, yeast, or mold. For these organisms, a D_M value of 200,000 rads or less was found, hence the 12 D_M value equals 2.4 million rads. Without relying on other agents to kill organisms, the following sterilization schedule is adhered to, on the basis of current knowledge:

(1) foods with pH values greater than 4.5 require 4.8 million rads.
(2) foods with pH values lower than 4.5 require 2.4 million rads.

The Food Product—Enzyme Destruction

The enzymes found in foods are generally more resistant to ionizing radiations than are micro-organisms. The "D_E" value is that dose which causes a 90 per cent reduction in enzyme activity. The destruction of enzymes with ionizing radiations is shown in a general manner in Fig. 103. It will be noted that the D_E value is much higher for enzymes than the D_M for micro-organisms.

The degree of enzyme inactivation required in radiation-stabilized foods is of the same order of magnitude as in other methods of preservation. It has been found that reductions in the order of four D_E units reduces activity to acceptable levels. The D_E value is approximately 5 million rads, and four times this value is 20 million rads. At this level, great destruction in food quality can be anticipated. Supplementary methods of enzyme inactivation are, therefore, indicated.

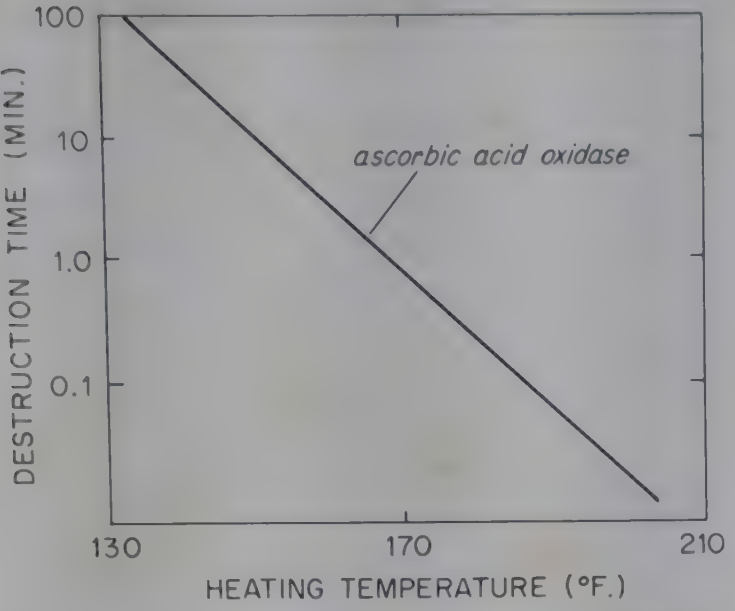
The Process for Food Stabilization

Satisfactory stabilization of a food solely with ionizing radiations will be obtained by applying either 12 D_M or 4 D_E levels of irradiation, whichever is greater.

Combination Treatment—
Heat Inactivation of Enzymes, Radiation Destruction of Micro-organisms

Because the enzymes are generally radiation resistant, it is desirable to inactivate them by means other than radiation. The well established heat sensitivity of enzymes offers an obvious solution; there may be many other methods of controlling enzymes. Enzymes are heat sensitive, and follow a pattern of destruction shown in Fig. 104.

If one D_E unit of destruction of enzymes can be obtained during the application of 12 D_M units of destruction of micro-organisms, the application of sufficient heat to bring about three D_E units reduction in enzyme activity should result in stabilization. In practice this effect has been used with good results. Actually, the application of less than four D_E units has been found to yield sufficient enzyme inactivation to permit several years of storage of irradiated foods without serious deterioration in food quality. Temperatures near 140°F., for periods upwards of 30 minutes with beef,



Courtesy of Esselen

Fig. 104. Destruction of Enzymes with Moist Heat (Ascorbic Acid Oxidase Shown)

have yielded acceptable levels of enzyme inhibition without sacrificing "medium-rare" appearance. The remaining enzyme destruction is accomplished when the beef is given 4.8 million rads.

Integration of the heat inactivation of enzymes with the destruction of enzymes with ionizing radiations is feasible. Combination treatments as above have been found to be useful in the destruction of organisms with mild heat treatments. The application of a million rads to putrefactive anaerobic spores reduces their moist heat sterilization requirements.

Combination Treatment—Antibiotic plus Radiation

The addition of antibiotics in low concentrations to foods has been found to reduce microbial populations, depending upon the spectrum of activity of the antibiotic (Cain *et al.* 1958). The addition of antibiotics to radiation-pasteurized foods appears to have merit (Huber 1958). By this means the storage life of "raw" commodities may be extended, especially with cool storage; great improvements in the distribution of perishables can be obtained. For example, the treatment of fresh fish with antibiotics, combined with irradiation, yields a cool storage life of fresh fish of up to two months compared to a week with aureomycin alone. Delivery of such "fresh" commodities, where economic advantages may be had, is an area which should receive attention.

Non-Heat Method for Controlling Enzymes in Meat

A pre-kill injection of the hormone epinephrine in animals has been found to depress the glucidic level of the tissues. After killing the animal, the pH value of the tissues remain over 7.0 for extended periods of time (upwards of a year). As a result the autolysis which usually occurs in stored meat is drastically curtailed. Such meat may be radiation stabilized without enzyme inactivation and stored for a year (and perhaps longer) at room temperature with little detectable autolysis. Sterile sections of animal flesh which have been so treated demonstrate this process (Radouco-Thomas 1958). A few milligrams of the hormone are given two hours before slaughter.

FOOD PROCESS DESIGN

Radiation Source Design

With the knowledge of the D_M and D_E values, and the thickness of the food package to be treated, the characteristics of the radiation source are defined. As noted earlier, the penetration power of gamma photons is so great that little obstacle is found in package thickness. The amount of radioactive gamma-emitting material required is dependent upon the dose demanded and the dwell (irradiation) time desired. Optimum design for the source is set by the time to give 12 D_M values. The radiation treatment should be given in 20 minutes or less, based on the generation time of micro-organisms. This demands rather large irradiator dimensions in terms of curies, leading to compromise with reality.

Unlike gamma photons, electrons have limited penetrating power. This poses certain design problems for accelerators. The energy requirement of the electron beam must take into account the thickness of the food packet. Each Mev of electrons can be expected to penetrate approximately one-fifth of an inch. A five Mev unit will penetrate one inch. A six-inch thick packet will require a 30 Mev beam if a single exposure is used. Bombardment of the food packet from opposite sides in a jaw-like apparatus, could reduce the energy for penetration to one-half, i.e., 15 Mev. But this energy level induces radioactivity in many elements.

Inasmuch as the killing effect of gamma photons and beta particles appear to be equal for equal doses, this factor need not be introduced into the discussion (Goldblith and Proctor 1954). The induced radioactivity found in high energy electron bombardment, especially over 25 Mev, may introduce a limit to packet size (Hannan 1956).

In radiation source design, two factors must be considered. First, the dose rate of the source must be compatible with the dose requirements for sterilization. Sources with dose rates of a few thousand rads per hour obviously involve such prolonged dwell times that product deterioration can be expected due solely to long term of the exposure. If doses near 5 million rads and exposure times of 20 minutes are desired, it is obvious that a radiation source permitting the absorption of 15 million rads per hour in the food is required.

Low Level Radiation Treatments

While sterilization of foods with ionizing radiations has usefulness in food preservation, processes yielding less than sterile food may also prove to have enormous advantages.

Low level or pasteurization radiation treatment of foods may be used to prolong the storage and distribution pattern for perishable commodities. Included are fruits, vegetables, meats, fish, poultry, and prepared foods for dinner tables.

The level of irradiation used is related to the degree of microbial and/or parasite destruction found useful, and of economic and/or public health significance.

Storage and therefore distribution patterns may be altered, and the areas in which foods are produced in the future will have greater freedom in marketing products with improving technologies.

The storage and distribution life of fresh foods can be extended by a factor of 5 to 10, using radiation treatments of a million rads or less. Furthermore, most packaging materials suitable for cool storage of perishable foods are unaltered functionally with such low levels of ionizing radiations (see Table 65). Costs for irradiation are at least one fifth less than those for sterilizing treatments, and even the latter is economically feasible.

Process and Product Specifications

With the development of consumer demand for radiation-treated foods will also come new concepts of process and product specifications. While many of the characteristics ordinarily used to describe food will be applicable to irradiated items, there will be differences between the specifications for radiation and conventional food preservation processes, as great as those between the specifications for frozen and dehydrated foods. It is useful to discuss the process and product specifications for various classes of radiation stabilized foods which now can be formulated on the basis of available information.

Process for Radiation Sprout Inhibited White Potatoes

The world production of potatoes is estimated to be 400 billion pounds annually. Under usual conditions, sprouting may occur about six weeks if unrefrigerated following harvest.

Potatoes for irradiation must be clean, well cured at the time of harvest, and free from diseases and bruises, e.g., in accordance with the require-

TABLE 65

THE COMPATIBILITY OF VARIOUS CONTAINERS WITH THE RADIATION PROCESS AND PROCESSING EQUIPMENT[1]

	Cans (with Enamels)		Semi-rigid Aluminum Dishes	Plastic Bag (Foil Lamination) in Carton					Wax Dip Coating
	Tinplate	Aluminum		Scotchpak (4.5 mil)	Mylar (2 mil)	Polystyrene (2 mil)	Nylon (2 mil)	Polyvinylchloride (2 mil)	
Radiation Process									
Change in physical properties (−10 per cent max)	+	+	+	+	+	+	+	+	+
Development of off-odors and flavors	+	?	?	−	+	−	?	−	?
Extractive substances	?	?	?	?	?	?	?	?	?
Manufacturing Process									
Manufacturing equipment	+	+	+	+	+	+	+	+	+
Filling equipment	+	+	+	+	+	+	+	+	+
Sealability	+	+	+	+	−	−	−	+	+
Sealing equipment	+	+	−	+	−	−	−	+	+
Availability in desired shape and size	+	+	?	+	+	+	+	+	+

---

[1] From: Tripp, G. Personal communication.
+ satisfactory; − unsatisfactory; ? unknown or borderline.

ments used to establish the U.S.D.A. standards for Grade No. 1 tubers (Workman 1959). Thick skinned tubers of the Russet Burbank (Idaho) type are found most satisfactory although other varieties such as (Maine) Katahdin and Irish Cobbler are also suitable. The temperature of the potatoes during irradiation should be below 70°F. Tubers are irradiated in such a manner that the dose absorbed by each tuber is not less than 6,000 rads nor more than 9,000 rads.

Radiation dosimetry is an essential new feature in this method of processing. Ferrous sulfate dosimeters discussed earlier are suitable for potato irradiation systems, and the dosimeter solution can be contained in small bottles, approximately $^3/_4$ inch in diameter and two inches in height. One dosimeter should be placed in every 50 standard boxes of potato tubers being irradiated. Health physics procedures complying with the Atomic Energy Commission and the National Bureau of Standards must be maintained at all times in the U.S.A. All treated potatoes must conform to the provisions of the Federal Food, Drug, and Cosmetic Act, the processing plant must obviously be maintained in sanitary condition, and the potatoes must be prepared and processed with a minimum delay between the various stages of processing. Following irradiation, the dosimeters are evaluated with a spectrophotometer, using a wave length of 3,020 Angstrom units (Hine and Brownell 1956). The calculated dose received by the potatoes should fall within the limits specified above. Those potatoes receiving less irradiation will sprout if stored at temperatures above 50°F., and those potatoes receiving excessive radiation will perform less satisfactorily in storage. Storage of potatoes should be at 41°F. and 85 per cent relative humidity. Potatoes can be packaged in 50-pound crates or in smaller consumer units. A minimum of handling of potato tubers is desired, and if potatoes can be packaged in the consumer unit, for example 10-pound string web or plastic bags, rehandling will be reduced to a minimum. For short term storage of potatoes at room temperature, approximately 20 per cent higher dosages should be given (Ellis *et al.* 1958).

Process for Insect Deinfestation of White Flour by Irradiation

All-purpose flour to be irradiated should be milled of clean and sound wheats of the classes such as those defined in the Handbook of Official Grain Standards of the United States for wheat. The grain should be free from smut, ergot, weed seeds, and foreign material. The bleaching agent used should produce the most beneficial effect on the flour with respect to color and bread-baking qualities, and the required enriching ingredients and other optional ingredients ordinarily added to flour may be present. Adjustment of the additives to compensate for their radiation destruction should be considered. The flour should have a moisture content of less than 14 per cent and an ash content of approximately 0.4 per cent. Flour used for irradiation should have a natural wheat flour taste and odor and be free from rancid, bitter, or musty tastes. Flour may be packaged in five or ten-pound paper or cloth bags which are sealed. Container thickness or package thickness must be considered with all products, and the radiation process for flour must give each package and its contents not less than 40,000 rads nor more than 60,000 rads (Baker *et al.* 1953). It is thought that a minimum average dose rate of 25,000 rads per hour is required for flour. Temperature of radiation should be below 75°F. The dosimeters for this range of radiation dose which have been found suitable are the ferrous sulfate dosimeters discussed above (J. Weiss 1952). A simulated package is constructed to hold the dosimeters, and the package design should be that of the packages of flour being treated. The dosimeter test container, as it may now be called, is then processed along with the flour. With a continuous flow of irradiated flour products through the radiation source, a test container for every 500 bags being processed should be considered. When exiting from the radiation source, no dosimeter should indicate less than 40,000 rads or more than 60,000 rads and assurance must be available that each package has in fact received the minimum dose. The finished flour must be white and free from abnormal odors and flavors. Health physics procedures and compliance with the provisions of the Federal Food, Drug, and Cosmetic Act must be maintained at all times in the U.S.A. The ferrous sulfate dosimeters following irradiation are evaluated with a spectrophotometer at a wave length of 3,020 Angstrom units. Individual sacks or bags of flour usually contained in a larger unit, commonly of a multi-walled paper construction. The overpack should be treated for insect-resistance in accordance with present commercial practices.

Process for Radiation-Pasteurized Plant Tissues (Fruits)

Only clean fruits free from disease and unbruised should be considered for irradiation. Fruits which are slightly less than fully ripened may be processed, as the radiation treatment will only temporarily interrupt normal ripening processes (Burns and Desrosier 1956). Fruits should be packaged to prevent reinoculation by air borne or accidental contamination. Packaging requirements are product specific and suitable materials are available commercially which have desirable carbon dioxide, oxygen, and moisture-vapor-transmission qualities. Reference should be made to Tables 45–50.

Radiation pasteurization of fruits is obtained with doses ranging from 150,000 rads to 180,000 rads, although this is dependent on the type of fruit and the length of storage life desired. Dosimetry is accomplished with ferrous sulfate or cobalt glass. Using cobalt glass, rectangles are made approximately 6 mm. by 15 mm. by 1.5 mm. in size, or slightly larger (see Chapter 7). When using glass dosimeters, the optical density is first recorded using a spectrophotometer operating at a wave length of 5,300 Angstrom units (Schulman *et al* 1955). A test container comparable to those being filled with food is prepared in which a series of cobalt glass dosimeters are located perpendicular to the largest dimension of the package. The dosimeters are positioned in containers as discussed above. Processing plant conditions and health safety precautions are similar to those for other foods. Storage of radiation pasteurized fruits must be under refrigeration to retard natural physiological aging processes, as the fruits retain their living features. Storage temperatures less than 40°F. are recommended, although this is again product specific, see references listed at the end of this chapter. Relative humidities between 85 and 90 per cent are required.

Process for Radiation Pasteurized Animal Flesh (Sliced Bacon)

Bacon prepared with good commercial practices from slabs ranging near ten pounds must be stored under refrigeration prior to being irradiated. Within an hour after slicing and packaging, the packaged bacon slices should be irradiated. If the packaged meat cannot be processed within an hour it must be held at 32°F. or frozen The packaged sliced bacon must be prepared in the U.S.A. under the supervision of the Meat Inspection Division of the U.S.D.A. and should be marked "inspected and passed" in accordance with existing regulations. Suitably packaged bacon is then exposed to a field of radiation to absorb a dose of not less than 800,000 rads nor more than 1,200,000 rads (Brownell *et al.* 1955). Radiation sources preferably should be capable of administering the required dose in less than 20 minutes. Temperatures within the food irradiation chamber should not exceed 75°F. The radiation source should be calibrated to indicate that induced activity does not exceed specified limits, for example, those established by the National Bureau of Standards Handbook #52 in the U.S.A. Dosimetry in this case will be undertaken with ceric sulfate as described previously. Cobalt glass could also be used (see also chapter 7). Small glass bottles of approximately one inch by three inches in dimensions are filled with the dosimeter solution and sealed. A simulated package of sliced bacon is then prepared in which several dosimeters are placed; the dosimeters may be held in place by agar, for example. With a continuous flow irradiation process a dosimeter container should occur at least once every 500 packages of sliced bacon. Following processing, the dosimeters are evaluated and none should indicate less than 800,000 rads or more than 1,200,000 rads. The irradiated sliced packaged bacon is stored at 32°F. The processed bacon should have white fat and the lean should have a cured color, and be free from abnormal flavors and odors. Slightly stronger curing and smoking practices are required (Erdman and Watts 1959).

Process for Radiation-Sterilized Meat (Chicken), Fish, and Vegetables

Chickens should be freshly killed, more than eight weeks old, and ranging in weight from $2^1/_2$–3 pounds prior to cutting into parts. Chickens must be free from evidence of disease or conditions which would render them unwholesome, and be processed in a sanitary manner. Individual chicken parts should not evidence having been frozen. Bruised parts and blood clots and other defects in chickens must be eliminated. Fresh chicken pieces must be enzyme inactivated, which is accomplished by exposing the flesh on a wire mesh belt which passes through a saturated steam atmosphere. Each piece should be free from direct contact with other pieces while being heated, and the pieces are cooked until they reach an internal temperature of not less than 165° nor more than 175°F. Immediately upon reaching this temperature, the flesh should be cooled to at least 50°F in less than one hour. When the chicken flesh has reached 50°F. or lower, the parts are packaged in containers of suitable size and sealed under vacuum. Within an hour following sealing, the packaged chicken parts should have been exposed to radiation. Meat, to be radiation-stabilized, must be exposed to a radiation source which permits the absorption in each container of not less than 4.8 megarads nor more than 5.2 megarads. The radiation source ideally should be capable of delivering this dose in approximately one hour and the temperature during irradiation should not exceed 75°F. Dosimetry measurements are made with ceric sulfate previously discussed. The vials of ceric sulfate solution are placed in containers similar to those containing the meat being irradiated. As an absolute minimum, at least one container containing dosimeters should be processed for every 500 cans of meat being sterilized. Preferably, it is good practice to have dosimeters located in the first, tenth, hundredth, and every hundredth unit thereafter, for greater control. Dosimeters should remain with each lot being processed during the period of irradiation. Following irradiation no dosimeter should indicate less than 4.8 megarads nor more than 5.2 megarads. The radiation-stabilized chicken has then received a heat treatment for enzyme inactivation, been sealed in a suitable container (usually tin plated steel cans) in a high vacuum (25 inches) and radiation sterilized. The surface color of the processed chicken flesh will now range from a very light pink to the characteristic gray-brown color of cooked chicken. When a freshly cut surface is viewed, it may be a slight pink color which, when exposed to the air, will fade to the characteristic color of cooked chicken. Color of the skin should remain a pale yellow. A four ounce packet of activated carbon in a suitable spill proof package should be included in each ten-pound container at the time of filling. All chicken used in the preparation of radiation stabilized chicken should be processed under the supervision of the Poultry Division of the U.S.D.A. in accordance with regulations governing poultry inspection in the U.S.A. Naturally, the process and the plant must be operated in such a way that sanitary products result. Chicken so processed after storage for 90 days at room temperature should be free from abnormal odors and flavors (see Fig. 100, page 309). The commercially available 0.25 pound electrolytic tin-plated steel cans with an inside enamel for chicken is available commercially from can manufacturers. Scotchpak, a laminated Mylar filmed flexible container is also satisfactory. A problem develops when irradiating food in cans under water. A six by seven inch cylindrical can containing a vacuum ranging from 20–25 inches when irradiated, for example, under 18 feet of water, will buckle. It is necessary to reduce the vacuum to between 12–16 inches, or alter the water head by one means or another.

Summary of Applications of Radiation to Food

It is seen from the above that the radiation stabilization of foods involves a number of considerations. There are four large categories of radiation treatment: sprout inhibition, insect de-infestation, pasteurization, and sterilization (Table 66). Sprout inhibition is seen to be best obtained when doses of 7,500 rads are given followed by storage of the tubers at 41°F. and 85 per cent relative humidity. Insect de-infestation is seen to be accomplished with 50,000 rads and provisions must be made to prevent reinfestation of the commodities by suitable packaging. The moisture content of the material to be de-infested is important; mold growth occurs in high moisture foods. Radiation-pasteurization of animal tissue is seen to be accomplished at 1,000,000 rads in suitably packaged material, followed by refrigerated storage. Most conventional packaging materials are serviceable for such treatments, but it must be recalled that the commodities so treated will not be sterile and cannot be stored safely at room temperature for prolonged periods of time. They require storage near 32°F. for prolonged storage lives, upwards of five times that normally obtained. Fruit pasteurization can be obtained with 150,000 rads with suitably packaged fruits subsequently cold stored. Radiation stabilization has three considerations, one, enzyme inactivation, accomplished by heating, two, destruction of micro-organisms, accomplished

TABLE 66

SUMMARY OF FOOD IRRADIATION PRODUCT AND PROCESS SPECIFICATIONS

Product	Process	Dose (rads)	Dosimeter	Package Requirements	Storage Temperature	Estimated Useful Storage Life
Potatoes	Sprout Inhibition	7,500	ferrous sulfate	stored in open containers or in porous containers	41 °F., 85% RH. 68 °F.	2 yrs. or more 10 mos. or less
Flour	Insect Destruction	50,000	ferrous sulfate	sealed paper or cloth bags, and overwrapped to prevent reinfestation	room temp. 40 °F.	2 yrs. or more 5 yrs. or more
Berries	Pasteurization (mold destruction)	150,000	cobalt glass	sealed in oxygen, CO_2 permeable film	34 °F.	21 days or more
Sliced Meat and Fish	Pasteurization (bacteria, yeast, mold, parasites, insects)	1,000,000	ceric sulfate	sealed in gas proof container	32 °F.	60 days or more
Animal, Fish, and Vegetable Tissue	Heat inactivation of enzymes (165 °F.). Radiation sterilization	4,800,000	ceric sulfate	vacuum sealed in durable container (tin can) with odor scavenger	room temp. 32 °F.	2 yrs. or more 5 yrs.
Fruits	Heat inactivation of enzymes (165 °F.). Radiation sterilization	2,400,000	ceric sulfate	vacuum sealed in durable container (tin can) with odor scavenger	room temp. 32 °F.	2 yrs. or more 5 yrs.

by the irradiation treatment, and three, prevention of reinfection, accomplished by packaging (Morgan and Siu 1957). The sterilizing dose presently is established from studies with *Clostridium botulinum* and is 4.8 megarads. Assurance must be had that all portions of all containers to be sterilized have in fact received this minimum dose to safeguard the public health. Suitable packaging must be included in radiation sterilization product and process specifications.

Containers found suitable for irradiation to date include the conventional tin-plated can, enamelled on interior surfaces in accordance with specific product requirements, and are available commercially at the present time. Aluminum and flexible packaging materials are also available. Most conventional flexible films, however, fail when exposed to more than two megarads, due to odor developments from irradiation of the films.

Radiation source technology must be applied in such a way that induced activity is at a minimum. Foods processed in accordance with present technology yield products which have induced activity far less than the amounts commonly found in water and foodstuffs in many localities. In fact, due to the wide range in radioactivity found in foods grown in various geographical areas of the world it may be anticipated that future legislation will establish upper limits in the radioisotope content of foods, whether irradiated or not.

BIBLIOGRAPHY

ALEXANDER, F., and FOX, M. 1952. Mode of action of agent giving protection from radiation. Nature *170*, 1022–1023.

ASTRACK, A. 1952. Effect of high intensity electron burst upon various vegetable and fish oils. Food Research *17*, 571–573.

BAKER, V. W., TABOADO, O., and WIANT, D. E. 1953. Lethal effect of electrons on insects infesting wheat and flour. Agr. Eng. *34*, 755–758.

BARRON, E. S. G., and DICKMAN, S. 1949. Studies on the mechanism of action of ionizing radiation. II. Inhibition of –SH enzymes by alpha, beta, and gamma rays. J. Gen. Physiol. *32*, 395–405.

BATZER, O. F., and DOTY, D. M. 1955. Nature of undesirable odors formed by gamma irradiation of beef. J. Agr. Food Chem. *3*, 64–68.

BELLAMY, W. D., and LAWTON, E. J. 1954. Problems in using high voltage electrons for sterilization. Nucleonics *12*, No. 4, 54–57.

BRASCH, A., and HUBER, W. 1947. Ultra-short application time of penetrating electrons: a tool for sterilization and preservation of foods in the raw state. Science *105*, 112–117.

BROWNELL, L. E., KEMPE, L. L., DENNIS, R. C., and GRAIKOSKI, J. J. 1955. Gamma rays: the hope of researchers to lengthen refrigerated life of fresh foods. Refrig. Eng. *63*, 42–47.

BURNS, E. E., and DESROSIER, N. W. 1956. Maturation changes in tomato fruits induced by ionizing radiations. Food Technol. *11*, 313–316.

CAIN, R. F., ANDERSON, A. W., and MALASPINA, A. S. 1958. Effect of radiation on antibiotic treated meats. Food Technol. *12,* 582–584.

CLIFCORN, L. E. 1955. The food industry's attitude toward radiation sterilization. Nucleonics *13,* No. 1, 39–41.

CORK, J. M. 1951. Radioactivity and Nuclear Physics. D. Van Nostrand Co., Inc., New York, N. Y.

CREAN, L. E., ISAACS, P. J., WEISS, G. J., and FAHROE, F. 1953. Application of isotopic source to food and drug sterilization. Nucleonics *11,* No. 12, 32–37.

DALE, W. M. 1940. The effects of X-rays on enzymes. Biochem. J. *34,* 1367–1373.

DALE, W. M. 1952. The indirect action of ionizing radiations on aqueous solutions and its dependence on the chemical structure of the substrate. J. Cellular Comp. Physiol. *39,* 39–55.

DESROSIER, N. W. 1958. Food irradiation research and its practical application in Europe and the U.S.A. Atoms for Peace Mission to Europe. E.P.A.-O.E.E.C. meeting on the application of atomic science in agriculture and food. Paris, France, July, 1958.

DESROSIER, N. W., *et al.* 1958. Progress and current status, 1954–1958, and future outlook. QM. Corps Radiation Preservation of Foods Project. QM. Food and Container Institute. Chicago, Ill.

DESROSIER, N. W. 1959. A challenge to the food industry of the U.S.A. in the peaceful use of atomic energy. Food Processing *19,* No. 2.

DESROSIER, N. W. 1959. The Technology of Food Preservation. Avi Publ. Co., Westport, Conn.

DESROSIER, N. W., MCARDLE, F. J., HOLLENDER, H. A., and MARION, W. W. 1955. What cathode rays do to milk and eggs. Food Eng. *26,* 78–79.

DEWEY, D. R. 1954. Electron linear accelerators for radiation processing. Nucleonics *12,* No. 12, 40–41.

DILLON, L. G., BURRIS, L., and RODGERS, W. A. 1954. Fission product kilocurie source: calculations of radiation intensity. Nucleonics *12,* No. 5, 14–16.

DOTY, D. M., and WACHTER, J. P. 1955. Radiation sterilization: influence of gamma radiation on proteolytic enzyme activity of beef muscle. J. Agr. Food Chem. *3,* 61–64.

DUFFY, D. 1953. Fission product potential of commercial reactors and their processes. Nucleonics *11,* No. 10, 8–11.

EDE, A. J. 1950. The electronic sterilization of foods. Food Sci. Abstr. *22,* 27–46.

ELLIS, N. K., *et al.* 1958. The feasibility of utilizing ionizing radiation to increase the storage life of potatoes. Final Rept. Proj. S-557. QM. Food and Container Institute. Chicago, Ill.

ERDMAN, A. D., and WATTS, B. M. 1957. Radiation preservation of meats. Food Technol. *11,* 349–353.

FIELDS, M. L. 1959. Effect of cathode rays on food spoilage fungi. Ph.D. Dissertation, Purdue University, Lafayette, Ind.

FRICKE, H. 1952. Effect of ionizing radiations on protein denaturation. Nature *159,* 965–966.

GLASSTONE, S. 1950. Sourcebook on Atomic Energy. D. Van Nostrand Co. Inc., New York, N. Y.

Goldblith, S. A., and Proctor, B. E. 1954. Relative merits of beta and gamma radiations from the standpoint of their application for sterilizing purposes. Nucleonics *12*, No. 2, 32–34.

Goldblith, S. A., and Proctor, B. E. 1952. Evaluation of food sterilization efficiency. Nucleonics *10*, No. 9, 28–29.

Goldblith, S. A., and Proctor, B. E. 1949. Effect of high voltage cathode rays on vitamins (riboflavin and carotene). Nucleonics 5, No. 2, 50–58.

Goldblith, S. A., Proctor, B. E., and Hemmerle, O. A. 1952. Evaluation of food irradiation procedures. Quantitative chemical measurements utilizing high energy cathode rays. Ind. Eng. Chem. *44*, 310–314.

Gomberg, H. J., Gould, S. E., and Bethell, F. H. 1954. Control of trichinosis by gamma irradiation of pork. J. Am. Med. Assoc. *154*, 654–658.

Goodman, C. 1949. The Science and Engineering of Nuclear Power. Addison Wesley Press, Cambridge, Mass.

Goodman, L. S., and Gilman, A. 1955. Toxicology. Macmillan Co., New York, N. Y.

Gowen, J. W. 1940. Inactivation of tobacco mosaic virus by X-rays. Proc. Natl. Acad. Sci. *26*, 8–11.

Goresline, H. E., and Desrosier, N. W. 1959. Preservation of foods by irradiation. Am. J. Public Health *49*, 488–493.

Hannan, R. S. 1954. Preservation of food with ionizing radiations. Food Sci. Abs. *26*, 121–126.

Hannan, R. S. 1956. Science and Technology of Food Preservation by Ionizing Radiations. Chemical Publ. Co., New York, N. Y.

Haurowitz, F., and Tremer, A. 1949. The proteolytic cleavage of irradiated proteins. Enzymologia *13*, 229–231.

Hayner, J. A., and Proctor, B. E. 1953. Investigations relating to the possible use of atomic fission products for food sterilization. Food Technol. *7*, 6–10.

Hine G. G., and Brownell, G. L. 1956. Radiation Dosimetry. Academic Press, Troy, N. Y.

Huber, W., Brasch, A., and Waly, A. 1953. The effect of processing conditions on organoleptic changes in foodstuffs sterilized with high intensity electrons. Food Technol. *7*, 109–115.

International Commission on Radiology Units. 1951. Recommendations of International Commission. Radiology *56*, 117–119.

Jackson, J. M., and Benjamin, H. A. 1948. Sterilization of food. Ind. Eng. Chem. *40*, 2241–2246.

Kempe, L. L., Kraikoski, J. T., and Brownell, L. E., 1953. Ray sterilizing food wrappers. Food Eng. *25*, 55.

Kraybill, H. F. 1955. Nutritive effects on foods sterilized by ionizing radiations. Nutrition Rev. *13*, 193–195.

Latarjet, R., and Caldas, L. R. 1952. Restoration induced by catalase in irradiated microorganisms. J. Gen. Physiol. *35*, 455–470.

Lea, D. E., Smith, K. M., Holmes, B., and Markham, R. 1944. Direct and indirect actions of radiations on viruses and enzymes. Parasitology *36*, 110–115.

Lehman, A. J., and Lang, E. P. 1954. Evaluating the safety of radiation sterilized foods. Nucleonics *12*, No. 1, 52–54.

LEVIN, S. 1954. Radiological safety in cathode ray sterilization. Nucleonics *12*, No. 6, 54–57.

LOEDING, J. W., PETKUS, E. J., YASNI, G., and RODGER, W. A. 1954. Fission product kilocurie source: preparation, radiation intensity. Nucleonics *12*, No. 5, 14–17.

MATELES, R. I., and GOLDBLITH, S. A. 1958. Some effects of ionizing radiations on gelatin. Food Technol. *12*, 629–633.

MCARDLE, F. J., and DESROSIER, N. W. 1955. Influence of ionizing radiations upon some protein components of selected foods. Food Technol. *9*, 527–532.

MCARDLE, F. J., MARION, W. W., and DESROSIER, N. W. 1954. Cold sterilization of fresh shell eggs. Proc. Poultry Sci. Ann. Meeting, June, 1954.

MCARDLE, F. J., and NEHEMIAS, J. V. 1956. Effects of gamma radiation on the pectic constituents of fruits and vegetables. Food Technol. *10*, 599–601.

MCNAMARA, A. T., CONQUEST, V., COONEY, J. P., GALLOWAY, C. G., and MCLAUGHLIN, V. J. 1958. Leaders appraise research in food irradiation. U. S. Army QM. Food and Container Institute. Activities Rept. *10*, No. 2, 57–68.

MECKER, B. A., JR., and GROSS, R. E. 1951. Low temperature sterilization of organic tissue by high voltage cathode ray irradiation. Science *114*, 283–285.

MEINKE, W. W. 1954. Does irradiation induce radioactivity in foods? Nucleonics *12*, No. 10, 37–39.

MORGAN, B. H. 1955. Food radiation round up. Food Eng. *16*, 43–47.

MORGAN, B. H., and SIU, R. G. H. 1957. A study of ionizing radiations on individual foods, Chap. 26, Radiation Preservation of Food. U.S. Govt. Printing Office, Washington, D. C.

NICKERSON, J. T. R., GOLDBLITH, S. A., and PROCTOR, B. E. 1950. A comparison of chemical changes in mackerel tissues treated by ionizing radiations. Food Technol. *4*, 84–88.

NICKERSON, J. T. R., LOCKHART, E. E., PROCTOR, B. E., and LICCIARDELLO, J. J. 1954. Ionizing radiations for control of fish spoilage. Food Technol. *8*, 32–34.

NICKERSON, J. T. R., PROCTOR, B. E., and GOLDBLITH, S. A. 1953. Public health aspects of electronic food sterilization. Am. J. Public Health, *43*, 554–560.

O'MEARA, J. P. 1953. Radiation chemistry and sterilization of biological materials by ionizing radiations. Nucleonics *10*, No. 2, 19–23.

PEARSON, A. M., DAWSON, L. E., BRATZLER, L. J., and COSTILOW, J. G. 1958. The influence of short term high temperature storage with and without oxygen scavenger on the acceptability of precooked irradiated meat. Food Technol. *12*, 616–619.

POLING, C. E., WARNER, W. D., HUMBURG, F. R., REBER, E. F., URBAIN, W. M., and RIA, E. C. 1955. Growth, reproduction, and histopathology of rats fed beef irradiated with electrons. Food Research *20*, 193–214.

POLLARD, E., and FARRA, F., JR. 1950. Cyclotron bombardment of enzymes and viruses. Phys. Rev. *78*, 335–341.

POWELL, W. F., and POLLARD, E. 1953. Radiation sensitivity of enzymes in intact cells. Phys. Rev. *79*, 494–496.

PROCTOR, B. E., and GOLDBLITH, S. A. 1952. Prevention of side effects in sterilization of food and drugs by ionizing radiations. Nucleonics *10*, No. 4, 64–65.

PROCTOR, B. E., and GOLDBLITH, S. A. 1951. Electromagnetic radiation fundamentals and their applications in food technology. Advances in Food Research *3*, 119–126. Academic Press, New York.

PROCTOR, B. E., and GOLDBLITH, S. A. 1950. Oxidation reduction dyes as radiation indicators. Nucleonics *7*, No. 2, 83–84.

PROCTOR, B. E., GOLDBLITH, S. A., BATES, C. J., and HAMMERLE, O. A. 1952. Biochemical prevention of flavor and chemical changes in foods and tissues sterilized by ionizing radiations. Food Technol. *6*, 237–242.

PROCTOR, B. E., JOSLYN, R. P., NICKERSON, J. T. R., and LOCKHART, E. E. 1953. Elimination of Salmonella in whole egg powder by cathode ray irradiation of egg megma prior to drying. Food Technol. *7*, 291–296.

PROGRESS REPORT ON ATOMIC ENERGY RESEARCH. Hearings before the Committee on Research and Development of the Joint Committee on Atomic Energy, Congress of the United States, 4–8 June 1956. U. S. Govt. Printing Office, Washington, D. C.

RADOUCO-THOMAS, C. 1958. Meat irradiation in relation to other methods of preservation with special reference to enzymatic autolysis inhibition by adrenaline. European meeting on the use of ionizing radiations for food preservation, Atomic Energy Research Establishment, Harwell, Eng. Nov. 1958.

ROBINSON, R. F. 1954. Some fundamentals of radiation sterilization. Food Technol. *8*, 191–194.

SCHMITZ, J. V., and LOWTON, E. J. 1951. Initiation of vinyl polymerization by means of high energy electrons. Science *113*, 718–719.

SCHULMAN, J. H., KLICK, C. C., and RABIN, H. 1955. Measuring high doses by absorption changes in glass. Nucleonics *13*, No. 2, 30–33.

SCHULTZ, H. W., CAIN, R. F., NORDAN, H. C., and MORGAN, B. H. 1956. Concomitant use of radiation with other processing methods for meat. Food Technol. *10*, 233–235.

SCHWEIGERT, B. S., NEVIN, B. S., DOTY, C. F., and KRAYBILL, D. M. 1954. Cold sterilization of meat. Am. Meat Inst. Found. Pamphlet No. 10.

SHEFFNER, A. L., ADACHI, R., and SPECTOR, H. 1957. The effect of radiation processing upon the In Vitro digestibility and nutrient quality of proteins. Food Research *22*, 455–461.

SILOW, R. A. 1958. The potential contribution of atomic energy to developments in agriculture and related industries. Int. J. Applied Rad. and Isotopes. *3*, 257–280.

SPARROW, A. H., and CHRISTENSEN, E. 1954. Improved storage quality of potato tubers after exposure to cobalt-60 gammas. Nucleonics *12*, No. 8, 16–18.

STANFORD, RESEARCH INST. 1951. Industrial uses of radioactive fission products. Report to the A.E.C., Project 361.

STOSIC, D. P. 1958. Rapid aging of wine with ionizing radiations. Personal communication, Belgrade, Yugoslavia.

TRUMP, J. G., and VAN DE GRAAFF, R. 1948. Irradiations of biological materials by high energy X-rays and cathode rays. J. Applied Phys. *19*, 599–604.

TRUMP, J. G., WRIGHT, K. A., and CLARKE, A. M. 1950. Distribution of ionization in materials irradiated by two and three million volt cathode rays. J. Applied Phys. *21*, 345–349.

UNITED NATIONS. 1958. European Meeting on the Use of Ionizing Radiations for Food Preservation. Atomic Energy Research Establishment, Harwell, Eng. Nov. 1958.

U. S. ARMY QM. CORPS. 1957. Radiation Preservation of Foods. U. S. Govt. Printing Office, Washington, D. C.

U. S. ARMY QM. FOOD AND CONTAINER INSTITUTE. 1958. Food preservation by irradiation. Digest of conference at Gatlinburg, Tenn. Activities Rept. 9, No. 2, 92–102.

URBAIN, W. M. 1953. Facts about cold sterilization. Food Eng. 25, 120–124.

WEISS, J. 1952. Chemical dosimetry using ferrous and ceric sulfates. Nucleonics *10*, No. 7, 28–31.

WORKMAN, M. 1959. On the inhibition of sprouting in potatoes. Personal communication. Lafayette, Indiana.

CHAPTER 19

Industrial Radiation Technology

The industrial application of ionizing radiation is a rapidly expanding field (Aebersold 1959). While the broad category of the industrial role of radiation has been discussed elsewhere (Little 1959, Wallenstein 1959) it is useful to discuss selected applications of radiation technology. Non-food sterilization, for example, has actually been the first industrial application of radiation because of its ability to kill organisms.

Pharmaceutical Products

It has been found that irradiation has advantages which make it attractive to the pharmaceutical industry. Radiation sterilizes without heating, and this is extremely important with heat sensitive pharmaceutical preparations. In fact, in many cases, radiation is preferred as a means of sterilization. For most therapeutic uses, pharmaceutical products must be completely sterile. Those products destined for external use only may require but a pasteurization treatment. Ultra-violet light has been used as a radiation source to sterilize plasma, but processing with ultra-violet light is limited because of its inherent low penetrating power.

The procedure for sterilizing with penetrating ionizing radiations differs considerably from conventional methods because the material to be processed can be packaged in a non-sterile area and then sterilized in the consumer package itself. Thus, the material and the interior of the package are sterilized at the same time. This negates the requirement that aseptic conditions be maintained throughout an entire manufacturing operation. So long as the treated package remains undamaged, the possibility of contamination which occasionally occurs in aseptic packaging systems does not exist. Material to be irradiated must be packaged and this is usually a machine operated process. The packaged pharmaceutical preparations are then irradiated sufficiently to destroy the organisms present, yet irradiated below the level which would cause undesirable effects in the product. After irradiation, samplings of the sterilized products are evaluated in the laboratory to establish that sterilization is absolute. However, this acts only as a check on the dosimeter system and when more confidence is gained in dosimetry in industrial processing, such biological testing will be rigorous. The same types of tests on concentration and potency obviously are made with radiation-sterilized material as are made with heat-sterilized or chemically sterilized preparations.

One of the important features of radiation sterilization in sealed packages is not only the elimination of costly aseptic areas and procedures commonly found today, but furthermore, irradiation can be a continuous operation, rather than a batch process which is generally the situation today. Continuous operations require less labor and improve efficiency in plants. One intangible asset of radiation sterilization is that a preparation need not be touched, the entire process may be mechanical. Many products which cannot be sterilized by conventional techniques may now be sterilized by irradiation. Radiation offers greater freedom in packaging, a limiting factor when some other methods of sterilization are used; with gamma radiation, deep penetration of substances is possible. And, there are indications that radiation treatments can result in new and better pharmaceutical products, aside from the sterilization aspect, examples being found in radiation synthesis procedures.

Radiation sterilization in the pharmaceutical and medical supplies industries is particularly advantageous where existing methods of sterilization are unsatisfactory. Examples of these are found in sutures, bandages, antibiotics, and ointments (Burrill and Gale 1952).

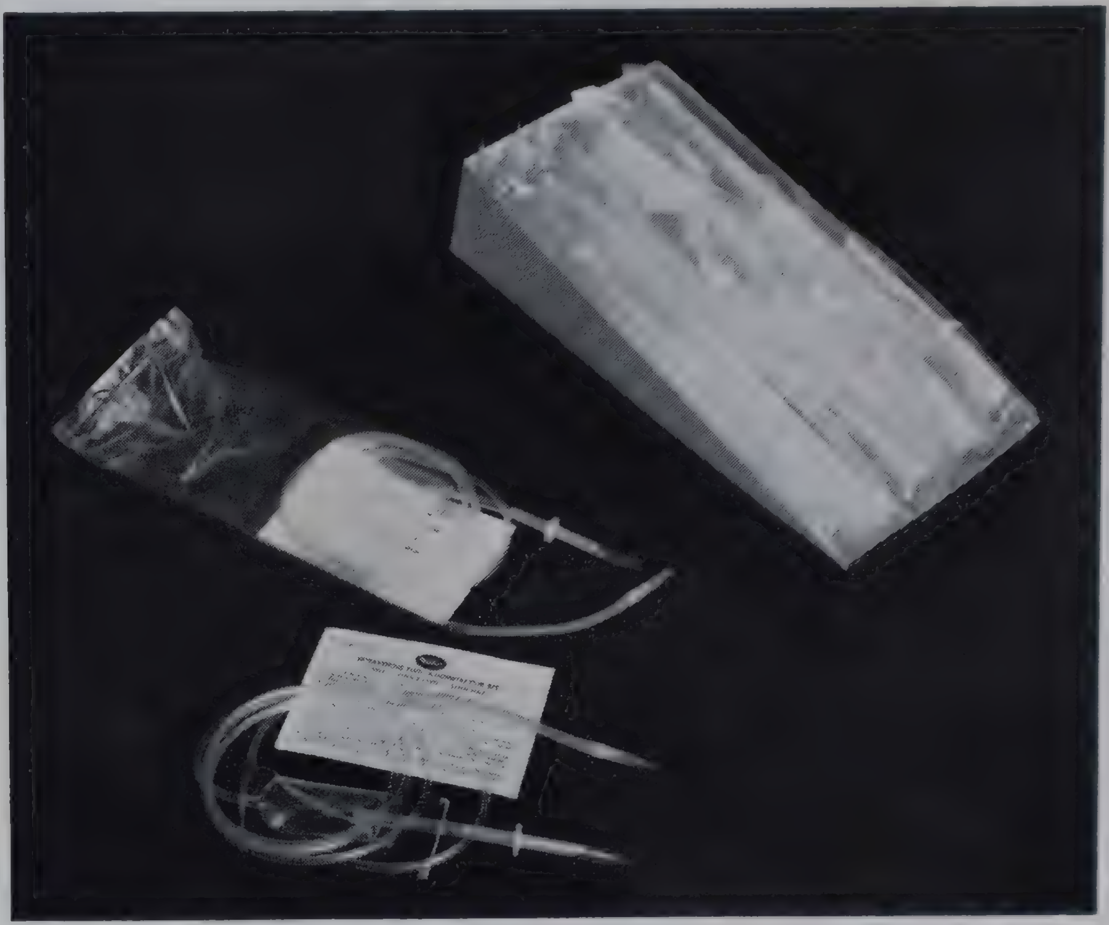
Radiation sterilized sutures were first marketed commercially in 1958 in the U. S. A. (Artandi and Van Winkle 1959). The conventional method of producing sterile sutures required sustained low temperature heat applications. For some sutures, chemical sterilization methods were used. However, the chemically sterilized and heat-sterilized products were different, and acute problems in production control occurred. After sterilization by either heat or chemical methods, processing still required skilled aseptic filling and packaging techniques, usually in glass containers. For the most part, processing and packaging were batch operations. In contrast, sutures that are to be ultimately radiation-treated are produced in a non-sterile environment with continuous flow procedures. The sutures do not have to be aseptically filled or sealed and the final container need not be glass. Many types of sutures are packaged in an organic liquid, and radiation sterilization eliminates this need. The ability to use plastic and metal foil-laminated packages has resulted in savings in handling, sterilization, packaging costs, labor requirements, processing and control, and has eliminated the inconvenience of glass breakage in hospital operating rooms. Furthermore, the resulting product is reported superior to competitive sutures and more economically produced. Radiation-sterilized sutures have a greater strength than heat-sterilized counterparts. While radiation cannot be used for all sutures, approximately 80 per cent of the 100,000,000 sutures produced annually in the United States will soon be radiation sterilized. It is anticipated that eventually the remaining production will also be radiation processed. Silk, catgut, and nylon are unaffected by the sterilizing radiation doses, while cotton sutures, as seen earlier, are degraded by ionizing radiations. Radiation treatment of sutures is an example where the initially higher cost of sterilization was offset by savings and advantages accruing elsewhere in the process from a better product to market advantages that are obtained through a more conveniently usable product.

Sutures were not the first pharmaceutical products which were irradiated. Ointments which were sterilized by radiation were marketed in 1955. One of these products contained the antibiotic neomycin and the hormone hydrocortisone. This was an eye ointment which, when irradiated, yielded single-use sterile units which reduced or eliminated the possibility of eye infections from contaminations by previous use of the ointment. This advantage was particularly useful in hospitals treating large numbers of patients. While in this particular instance, however, radiation sterilization has been discontinued, apparently for economic reasons, the economic factor may change in the future and again make the process.attractive.

Antibiotics are produced by fermentation under rigid conditions, isolations and crystallization of the antibiotic, sterilization of the crystals with gas, and packaging under aseptic conditions. Savings in the production of sterile antibiotics with irradiation are found in crystal sterilization and in packaging. It would appear that the commercial use of radiation with antibiotics is dependent upon having a lower process cost for producing antibiotics which can be satisfactorily irradiated. Some of the important antibiotics lose their potency due to the irradiation process. Improved technology may overcome even this difficulty.

Bandages, Bulk Medical Supplies

Cotton, gauze, and adhesive bandages, surgical dressings, and other bulk medical materials make up the largest volume of medical supplies now being sterilized by conventional methods. The annual market for these products is in excess of $100,000,000 in the U. S. A. alone. Approximately 50,000,000 pounds of bandages are sterilized by heat currently, and this figure is increasing annually, along with the increases in population. Currently, pallet loads of packaged bandages or bulk medical supplies are placed in compression chambers and given a pressurized steam treatment for a sufficient period of time to kill the micro-organisms present. Success is dependent upon sufficient penetration of steam into the package to provide moist heat conditions to destroy the organisms, dry heat being relatively ineffective. The use of gamma radiation for the sterilization of bulk materials such as bandages after packaging and prior to shipment to markets or to hospitals has striking advantages. The ability of lethal gamma radiation to penetrate pallet loads of materials is easily recognized. Furthermore, complete sterilization of even the most tightly packed containers is possible with gamma radiation, and nearly impossible with steam. The potential savings here are notable even in packaging materials and there is a wide freedom in the type of packaging materials which can be used in this application. There is an elimination of waste products, heat sensitive material being irreversibly damaged by over-exposure to steam, an inherent problem with conventional heat penetration. Radiation offers the advantage of continuous processing methods in comparison to the batch type in present use, leading to improved, more economical material handling and processing methods. An example of an intravenous fluid administration set sterilized with radiation is shown in Figs. 105 and 106.

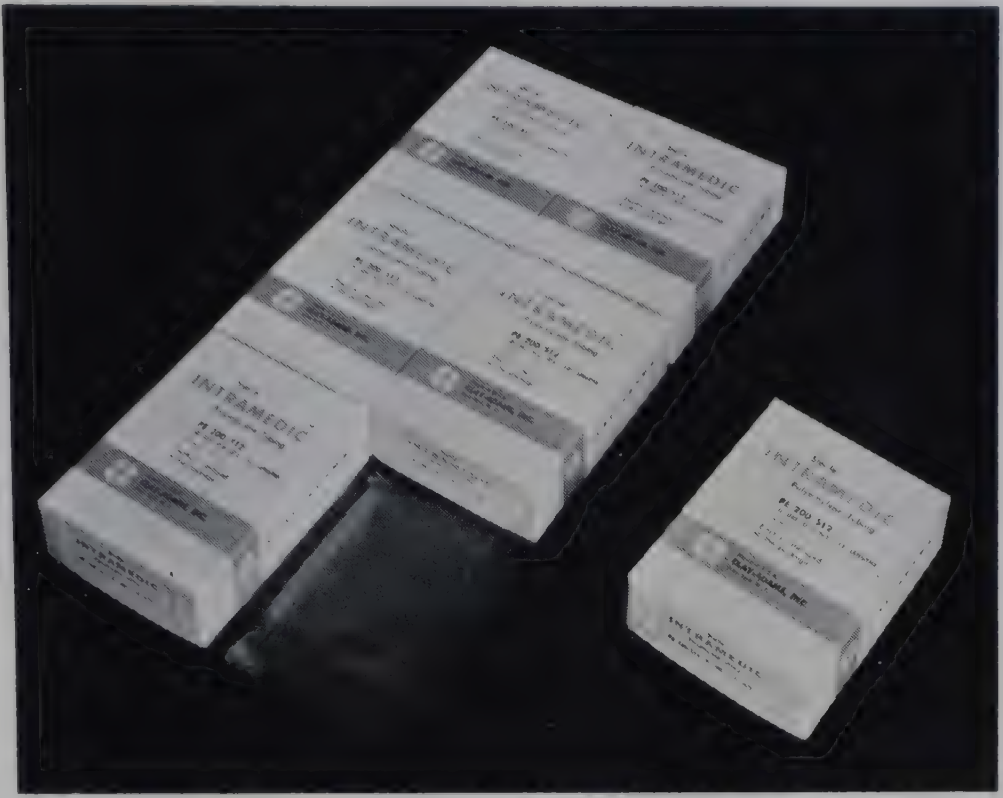


Courtesy of High Voltage Engineering Corp.

FIG. 105. RADIATION STERILIZED INTRAVENOUS FLUID ADMINISTRATOR SET

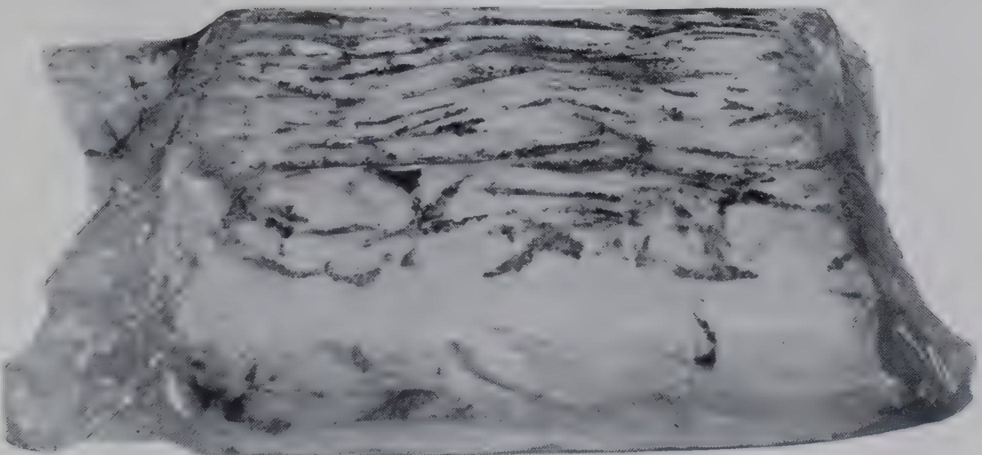
Improving Fibers, Leathers, and Wood

The molding of clothing, textiles, canvas, tenting, and leather products is a serious problem in warm and humid areas of the world. The ability of ionizing radiation to destroy molds has previously been discussed. The elimination of molds in packaged clothing (Fig. 107) is noteworthy.



Courtesy of High Voltage Engineering Corp.

Fig. 106. Commercially Produced Radiation Sterilized Tubing for Medical Applications



Courtesy of U.S. Army Quartermaster Corps

Fig. 107. Molding of Clothing can be Eliminated with Radiation Treatment of Packaged Material

The prevention of molding of leather shoes, for example, using suitable inexpensive packaging followed by low level irradiation has great promise in eliminating an important economic loss.

Recent reports indicate the use of radiation to improve the properties of natural textile fibers, wood, and leather. When used alone, some deleterious effects of radiation are found. However, radiation has been found useful in graft polymerization of natural fibers with monomeric molecules to achieve better chemical and physical properties of these natural materials. Wood impregnated with styrene, for example, has been found to have improved strength and improved moisture and swelling resistance when irradiated. Because of the large volume of natural fibers, wood and leather produced annually, their improvement by such techniques would seem a fruitful area of investigation.

Cellulose Degradation

As previously noted, the irradiation of cellulose molecules yields some sugar. Suggestions for degrading cellulose from wood or plant stocks to make suitable cattle feed have been made. The useful degradation of cellulose requires megarad doses and would obviously require low cost radiation sources to be economically feasible.

Upgrading Natural Oils for Industrial Uses

The irradiation of natural oils has been found to increase the double bonds present, which is an important characteristic in drying oils (Astrack 1952). The chlorination, oxidation, and polymerization of a variety of organic materials are equally noteworthy (Hills *et al.* 1959).

Soil Sterilization

Certain plant and animal diseases contaminate soil and reinfect crops and animals annually. Crop rotation and chemical treatments are the present control methods employed. Some recent work undertaken with irradiation of the top six inches of land with portable radiation source have had limited success; a dose of 15,000 rads administered to soil infested with potato nematodes does not destroy them. Larger doses could produce sterility in soil and, importantly, prevent the sprouting of weed seeds, but the practical means of carrying out such an irradiation at a reasonable cost requires further research and development. While certain advantages of soil irradiation seem attractive in terms of increased yields and disease control, many problems must be overcome before radiation sterilization of soil can compete with the conventional techniques.

Although previously discussed in several chapters, the following applications of ionizing radiations should be reviewed:

1. The destruction of parasites in water supplies, such as amoebic dysentery organisms, which are resistant to chlorine treatment could make substantial contributions to public health. Sewage disposal applications, including night soil control, would appear promising (Fowler, 1959).

2. Low level irradiation of meat animals for parasite control is within the grasp of present technology. Inexpensive control is possible with present knowledge and could be applied almost immediately in eliminating certain infections of man by those parasites transmitted through water and food supplies.

3. The control of insect populations destructive to man and his animals has found certain success. Sterilization of segments of screw-worm populations, leading to sterile off-spring has been widely applied in controlling screw-worm infestations of cattle in South America and in southern U.S.A. areas.

4. Low level irradiation of seeds has yielded interesting results. Crop production initially was found to be increased by a significant factor. These studies are at their earliest beginnings, but preliminary results are promising. This application is in addition to the useful radiation-induced mutations.

Medical Therapy

Almost immediately following the discovery of radioactivity in 1896, X-rays were applied to the treatment of malignant diseases. Next to surgery, radiation therapy is one of the principal means of cancer control. Radiation doses ranging from 6,000 to 9,000 rads are required, and cost is not a factor. Cobalt-60 radiation sources are presently in use in more than 100 hospitals in the U. S. A. alone for the treatment of various kinds of cancerous tissues. Cesium-137 sources have also been used for this purpose and it seems reasonable to believe that cesium sources will be important in this application in the future. This is an area expanding rapidly throughout the world.

Diagnostic Uses of Radiation

The use of radiography has been well established for several decades. Its use is now increasing not only in the diagnosis of acute illness, but also in preventive medicine, for example the X-ray examination of members of whole populations to detect tuberculosis and heart diseases. When emergency situations arise, electric power required for X-ray equipment is not always available. The need for portable, lightweight, flexible radiography units independent of electric power is fulfilled with radioisotope sources.

Industrial Radiography

One of the first techniques used in the non-destructive testing of materials employed radiation. X-ray machines, as well as radium, were used as industrial tools. Radiation sources for non-destructive testing of materials and structures have been employed with ships, aircraft, boilers, and missiles and the list of uses is growing. At the present time, it has been reported that there are more than 500 organizations using radiation soruces for radiographical purposes in the U. S. A. (Aebersold 1959).

Radioactive Isotopic Tracers

The use of radioisotopes in research on the basic mechanisms of life has found widespread application in laboratories throughout the world. The results obtained are already of great value. Examples are seen in the better utilization of fertilizers in agriculture. The use of radioactive carbon has yielded important information on the photosynthetic process itself and it is recalled that practically all agriculture and life are based on the photosynthetic process. Flight patterns of insects have been traced using radioisotopes and knowledge of plant and animal as well as human diseases has been greatly improved by the use of radioisotopes. At the present time, laboratories which have no radioisotope research facilities find great difficulty in maintaining themselves in a favorable position at the horizon of information being developed in many areas of research in the physical and biological sciences.

The use of radioisotopes in diagnosing human illnesses is also becoming of increased importance. The introduction of radioactive iodine with its subsequent concentration in a goiter, for example, is a well-established medical diagnostic technique. Such uses are increasing rapidly.

Chemical Processing

Radiation-induced chemical reactions were discussed in Chapter 4. The new freedom permitted by radiation chemistry is the great flexibility of obtaining reactive species in a variety of surroundings and over wider ranges of temperature and pressure. Furthermore, the very fact that the irradiation of a molecule can produce pronounced changes in a single step offers great promise in eliminating the commonly found numerous steps in chemical processing which are required to effect a particular chemical change (Wallenstein *et al* 1959). Perhaps the most dramatic possibility is that of creating chemical changes which are, at present, very difficult to carry out. It was seen earlier that in the final analysis the radiation part of radiation chemistry is indeed a new way of creating reactive chemical species. The chemistry part of radiation, as was discussed earlier, abides by the rules of formation and reaction of chemical species and the same set of rules apply that cover conventional chemical changes. In the polymer field, the most important feature of radiation application is in the formation of reactive free radicals and ions. The ability of high energy radiation to generate these species in a controlled manner has made radiation a powerful tool in polymer research and has opened many possibilities for process applications in polymer technology. Radiation is usefully employed in polymerization of monomers, crosslinking of polymers, degradation of polymers, and graft and block co-polymerization. Radiation applications in chemical reactions of possible industrial importance include the formation of lindane, an insecticide, from benzene and chlorine, the direct production of phenol from benzene, water, and oxygen, the direct fixation of nitrogen, the synthesis of ascorbic acid, and the formation of detergents from hydrocarbons and sulfur dioxide, to name but a few. Radiation is seen to have widespread application in oxidation as well as in substitution reactions and in controlled molecular rearrangements. Also of interest is the ability to desensitize milk protein for example and the rapid radiation aging of wines. The former is important in feeding children allergic to cow's milk; the latter may be useful in eliminating the aging cellars for wines, or increasing their capacities.

Synthesis of Sugar Using Radiation.—An example of the application of radiation in synthesis may be seen in the synthesis of sugar from water and carbon dioxide. The electrolysis or radiolysis of water yields hydrogen and oxygen gas. The radiolysis of carbon dioxide yields carbon monoxide and oxygen. Carbon monoxide plus hydrogen in the presence of the catalyst zinc chromate yields methyl alcohol, the process used commercially. The irradiation of methyl alcohol was shown earlier to yield two and three carbon polyhydroxy alcohols which then can be made to yield sugar. As shown earlier by Urey (1952), Miller and their coworkers, some of the essential amino acids may be produced by the irradiation of a mixture of hydrogen, ammonia, methane, hydrogen sulfide, and water. It is clear that the possibilities of the formation of intermediate carbon compounds of value offer an intriguing area of study.

The Role of Radiation in Other Areas

When a solid material is irradiated, some of the atoms in the material are displaced from their normal position in the solid lattice. Also, some of the electrons are excited to high energy levels or are caught in the so-called electron traps. These effects result in changes in the electrical, mechanical, and optical properties of solids. One result of electron excitation is the production of luminescence much in the same manner as in the every day electric fluorescent lamp. The production of completely self-contained light sources of very long life using radioisotopes offers attractive possibilities for specialized uses. Some promising commercial applications include railroad signal lamps for use in switch yards and non-electrified rural lines and emergency exit signs (Little 1959).

The use of radionuclides to produce heat, which in turn produces electricity has been the subject of much attention in many laboratories. Such devices which produce electricity by this means have potential for long performance, and absence of temperature, pressure, and acceleration effects (Barmat *et al.* 1959) which may make them significant in space flight.

Another consequence of atomic electron displacement in a solid is the change in catalytic properties which may result therefrom (Kohn and Taylor 1959). While the detailed nature and mechanism leading to these changes in properties are not well understood, in view of the usefulness of catalytic technology, the irradiation treatment of catalysts should have imporant tèchnological consequences. The ability to produce sterile enzymes is of equal note, taking advantage of the greater radiation resistance of enzymes over organisms, important in dissolving blood clots.

Another use of radioisotopes is in the field of chemical tracers in industrial processing (Hull 1955). Tracer techniques are finding wide applications in research and control, ranging from the measurement and/or control of thicknesses, fluid flow, liquid level, and other features of importance in industrial operations.

In summary, it can be said that the industrial uses of radiation are increasing annually and future applications are anticipated which will yield increased benefits. Considering the large number of laboratories and scientists concentrating in this area of activity throughout the world, it is not surprising to find new application areas appearing monthly in the current literature.

The field of high energy radiation applications is potentially of unlimited breadth. Those applications directly related to power production, mainly in the power reactors, are important in making possible the industrial uses of radiation.

The reactor yields a by-product of radioactive wastes. Radiation technology contributes to the successful exploitation of nuclear energy by improving the economics of power production through usefully employing its waste products.

BIBLIOGRAPHY

Aebersold, P. C. 1955. Importance of isotopes in technology and industry. Proc. Intern. Conf. Peaceful Uses of Atomic Energy *14*, 3–20. Geneva.

AEBERSOLD, P. C. 1959. The United States industrial radiation program. Intern. Atomic Energy Agency, Warsaw Conference: Application of Large Radiation Sources in Industry. Paper CW/S/57.

ARTANDI, C., and VAN WINKLE, W., JR. 1959. Electron beam sterilization of surgical sutures. Nucleonics *17*, No. 3, 86–90.

ASTRACK, A. 1952. Effect of high intensity electron bursts on various vegetable and fish oils. Nature *170*, 1022–1023.

ATOMIC INDUSTRIAL FORUM, INC. 1955. A growth survey of the atomic industry, 1955–1965. New York, N. Y.

BARMAT, M., ANDERSON, G. M., and BOLLMEIER, E. W. 1959. Electricity from radionuclides and thermoelectric conversion. Nucleonics, *17*, No. 5, 166–174.

BOPP, C. D., and SISMAN, O. 1955. How radiation changes polymer mechanical properties. Nucleonics *13*, No. 10, 51–55.

BROWNELL, L. E., *et al.* 1955. Utilization of gross fission products. Eng. Res. Institute, Univ. of Michigan, Ann Arbor, Michigan.

BRYNJOLFSSON, A., and HOLM, N. W. 1959. The cobalt-60 irradiation facility and gamma field at Rise, Denmark. Intern. Atomic Energy Agency, Warsaw Conference. Paper CW/Ipl/75.

BURRILL, E. A., and GALE, A. J. 1952. Electron beams sterilize food and drugs. Electronics *25*, 98–101.

BURTON, M. 1959. Elementary processes characteristic of radiation chemistry and possible industrial implications. Intern. Atomic Energy Agency, Warsaw Conference: Application of Large Radiation Sources in Industry. Paper CW/S/66.

CHRISTENSEN, C. J., JACOBSEN, A., KLINGER, Q., and ABEL, G. 1957. Dugway food irradiator uses 8 MTR fuel elements. Nucleonics *15*, No. 12, 94–95.

COLICHMAN, E. L., MALLON, P. J., and JARRETT, A. A. 1957. Gamma facility for use at high temperatures. Nucleonics *15*, No. 4, 115–118.

DESROSIER, N. W. 1958. Food irradiation research and its practical application in Europe and the United States. Project Rept. No. 396. E.P.A.-O.E.E.C 2 Rue Andre Pascal, Paris, France.

DESROSIER, N. W. 1959. The Technology of Food Preservation. Avi Publ. Co., Inc. Westport, Conn.

DOBO, J., SOMOGYI, M., and KISS, L. 1959. Synthesis of graft copolymers by small doses of irradiation. Intern. Atomic Energy Agency, Warsaw Conference. Paper CW/IIB/3.

EHRENKRANZ, F., and JEBE, E. H. 1956. Carbon-14 method of testing home laundering procedure. Nucleonics, *14*, No. 3, 96–101.

FAHNOE, F., CREAN, L., and KUPP, R. 1952. Industrial applications of gross fission products. U. S. Atomic Energy Commission Con. No. AT (30-1)-850. Germantown, Maryland.

FOWLER, E. B. 1959. Irradiating sewage might improve bacterial nutrients. Nucleonics *17*, No. 4, 140–142.

GEORGE, D. W., and GREGORY, J. N. 1959. Gamma radiation facilities at the Australian Atomic Energy Commission Research Establishment. Intern. Atomic Energy Agency, Warsaw Conference: Application of Large Radiation sources in Industry. Paper CW/Ipl/12.

GLASSTONE, S. 1958. Sourcebook on Atomic Energy. Second Ed. D. Van Nostrand and Co., New York, N. Y.

Hannan, R. S. 1956. Science and Technology of Food Preservation by Ionizing Radiations. Chemical Publ. Co., New York, N. Y.

Harlan, J. T., and Hart, E. J. 1959. Ceric dosimetry: accurate measurements at 10^8 rads. Nucleonics *17*, No. 8, 102–104.

Harteck, P., and Dondes, J. 1956. Producing chemicals with reactor irradiation. Nucleonics *14*, No. 7, 22–25.

Harteck, P., and Dondes, S. 1957. Glass fibers—a new form for reactor fuels. Nucleonics *15*, No. 8, 94–98.

Hills, P. R., Roberts, R., and Spindler, M. W. 1959. A comparison of the production of gammexane by ultra-violet and cobalt-60 gamma irradiation. Intern. Atomic Energy Agency, Warsaw Conference: Application of Large Radiation Sources in Industry. Paper CW/IIC/19.

Holmes, B. 1947. The inhibition of ribo- and thymo-nucleic acids synthesis in tumor tissues by irradiation with X-rays. Brit. J. Radiol. *20*, 450–453.

Huber, W. 1948. Sterilization of drugs. Electronics *21*, 74–79.

Hull, D. E. 1955. Using tracers in refinery control. Nucleonics *13*, No. 4, 18–21.

Intern. Atomic Energy Agency. 1959. Abstracts of papers for the conference on the application of large radiation sources in industry: Warsaw Conference. Vienna, Austria.

Jefferson, S., Murray, G. S., and Rogers, F. 1959. Gamma radiation sources of the English Technological Irradiation Group, Intern. Atomic Energy Agency, Warsaw Conference: Application of Large Radiation Sources in Industry. Paper CW/Ipl/87.

Johnston, W. H., Wallenstein, M. B., Rosenstock, H. M., and Desrosier, N. W. 1959. Industrial Role of Radiation. Research Rept. Con. No. AT(11-1)-650. U. S. Atomic Energy Commission, Office of Isotope Development, Germantown, Maryland.

Kohn, H. W., and Taylor, E. H. 1959. The effect of ionizing radiations on gamma Al_2O_3 as a catalyst for H_2-D_2 exchange. J. Phys. Chem. *63*, 500–505.

Kraybill, H. F. 1958. Desensitizing proteins for human consumption. Personal communication. Denver, Colo.

Kuhl, O. A., Sparrow, A. H., and Manowitz, B. 1955. Portable pilot plant for irradiating potatoes. Nucleonics *13*, No. 11, 128–129.

Leveque, P. 1959. Description of sources for irradiation. (in French). Intern. Atomic Energy Agency, Warsaw Conference: Application of Large Radiation Sources in Industry. Paper CW/Ipl/40.

Libby, W. F. 1957. Radioisotopes. Nucleonics *15*, No. 4, 118–121.

Little, A. D., Inc. 1959. Radiation: A Tool for Industry. Con. Rept., Office of Isotope Development, U. S. Atomic Energy Commission, Germantown, Maryland.

Manowitz, B., Kuhl, O. A., Galanter, L., Bretton, R. H., and Swickler, S. 1959. Experimental parametric study of large scale cobalt, sodium, and cesium slab irradiators. Intern. Atomic Energy Agency, Warsaw Conference. Paper CW/I/70.

Manowitz, B., and Richman, D. M. 1956. Economic future of fission products for radiation power. Nucleonics *14*, No. 6, 98–102.

MATSUMURA, S., FUJII, T., and KONDO, S. 1959. Cobalt-60 gamma irradiator and its use in plant genetics. Intern. Atomic Energy Agency, Warsaw Conference: Application of Large Radiation Sources in Industry. Paper CW/IVA/30.

MICHAELIS, M. 1959. Technology and economics of large radiation sources. Intern. Atomic Energy Agency, Warsaw Conference: Application of Large Radiation Sources in Industry. Paper CW/IIIP/71.

NATIONAL BUREAU OF STANDARDS. 1953. Maximum Permissible Amounts of Radioisotopes in the Human Body and Maximum Concentration in Air and Water. Handbook No. 52. U. S. Govt. Printing Office, Washington, D.C.

NUCLEONICS REPORT. 1956. How radiation affects materials. Nucleonics *14*, No. 9, 53–88.

NUCLEONICS REPORT. 1958. Understanding nuclear power costs. Nucleonics *16*, No. 1, 46–57.

NUCLEONICS REPORT. 1959. Food irradiation looks to low dose treatments. Nucleonics *17*, No. 10, 112–113.

NUCLEONICS REPORT. 1959. Measuring large radiation doses. Nucleonics *17*, No. 10, 57–76.

PRIMAK, W., and FUCHS, L. H. 1955. Nitrogen fixation in nuclear reactor. Nucleonics *13*, No. 3, 38–41.

PROCTOR, B. E., LOCKHART, E. E., and GOLDBLITH, S. A. 1954. The application of electronic treatments to the destruction of insects in packaged military rations and packaging materials. Final report, U. S. Army QM. Corps. Con. #DA-11-009-QM-19888. Chicago, Illinois.

PROGRESS REPORT ON ATOMIC ENERGY RESEARCH. 1956. Hearings before the Committee on Research and Development of the Joint Committee on Atomic Energy, Congress of the United States, 4–8 June. U. S. Govt. Printing Office, Washington, D. C.

RANFTL, J. W. 1959. Irradiated polyethylene—a case study in economics of production use of electron beam radiation. Intern. Atomic Energy Agency, Warsaw Conference: Application of Large Radiation Sources in Industry. Paper CW/IIIp/54.

RICE, F. G., COURTEMANCHE, R., and MASEFIELD, J. 1959. Preliminary design study of a mobile irradiator. Iitern. Atomic Energy Agency, Warsaw Conference: Application of Large Radiation Sources in Industry. Paper CW/I/20.

SCHMITZ, J. V., and LOWTON, E. J. 1951. Initiation of vinyl polymerization by means of high energy electrons. Science *113*, 718–719.

SILOW, R. A. 1958. The potential contribution of atomic energy to development in agriculture and related industries. Intern. J. Applied Radiation and Isotopes *3*, 257–280.

STANFORD RESEARCH INSTITUTE. 1951. Industrial uses of radioactive fission products. Report to the U. S. Atomic Energy Commission Project 361.

SIU, R. G. H. 1959. Current concept and status of radiation preservation of foods. Intern. Atomic Energy Agency, Warsaw Conference: Application of Large Radiation Sources in Industry. Paper CW/S/7.

SUN, K. H. 1955. A comparison of radiation sources and some typical applications in the chemical industry. Atomic Energy in Industry Conference. New York, N. Y. Oct. 26–28.

SYMPOSIUM ON THE INDUSTRIAL ROLE OF RADIATION. 1959. Georgia Institute of Technology, Atlanta, Georgia.

TAIMUTY, S. I., FOWLE, L. H., and PETERSON, D. L. 1959. Ceric dosimeter for routine use at 10^5 to 10^7 rads. Nucleonics *17*, No. 8, 103–107.

THOMAS, A. 1955. Nuclear batteries: types and possible uses. Nucleonics *13*, No. 11, 129–133.

UNITED NATIONS. 1958. Peaceful Uses of Atomic Energy. Proc. of the second Intern. Conference, Geneva, Switzerland. 33 volumes.

UREY, H. C. 1952. The Planets, Their Origin and Development. Yale University Press, New Haven, Conn.

UREY, H. C. 1952. The origin of the earth. Sci. Monthly *187*, No. 4, 53–60.

U. S. ARMY QM. CORPS. 1957. Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

UNITED STATES JOINT COMMITTEE ON ATOMIC ENERGY. 1956. Peaceful Uses of Atomic Energy. Rept. of the Panel on the Peaceful Uses of Atomic Energy. U. S. Govt. Printing Office, Washington, D. C.

VIDAL, P. 1959. Rapid aging of wines with low level irradiation. Personal communication. Lyon, France.

VIDAL, P. 1959. Improvement of yield and quality of crops by seed irradiation (in French). Industries Atomiques *3*, 77–84.

WALLENSTEIN, M. B., ROSENSTOCK, H. M., and JOHNSTON, W. H. 1959. Fundamental considerations of radiation technology. U.S.A.E.C. Contract Rept. AT(11-1)-650. Parts I, II.

CHAPTER 20

Radiation Facilities and Process Economics

Radiation sources for application in the fields of food, agriculture, and biology may vary substantially in design. Radiation source design, however, is beyond the scope of this book, and the reader is referred to the bibliography at the end of this chapter.

The earliest radiation sources of intensity sufficient to permit high dosages to be absorbed in substantial quantities of materials were of two types, electron machines and spent fuel elements from reactors. Both were discussed in Chapter 6.

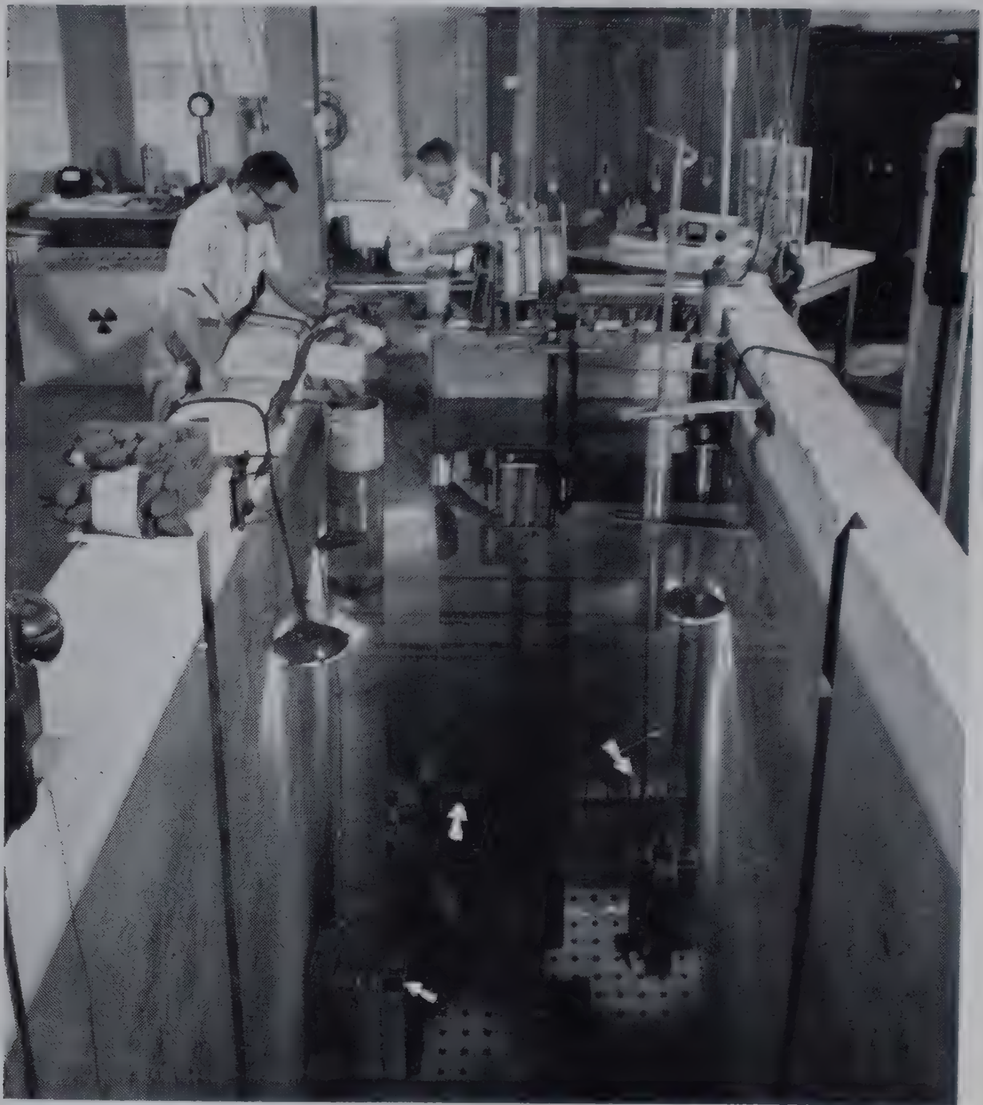
Early Radiation Facilities

One of the earliest radioactive sources with useful high gamma flux was established at the Materials Testing Reactor site at Arco, Idaho, which was discussed in Chapter 6. The gamma facility is shown in Fig. 108. Spent fuel elements from the neighboring reactor were placed beneath about 16 feet of water in a canal for shielding. Since opportunities exist for contamination of the canal water, stainless steel tubes were constructed which had no direct access to the canal water itself. Such isolated tubes could be constantly purged with fresh uncontaminated water. Material to be irradiated could be lowered within the tubes into the area of the fuel elements, themselves positioned around and outside the tube base, shown in the figure. Dose rates with spent fuel elements a few days after being removed from the reactor core amounted to some 500,000 rads per hour and more. The operation was a batch process.

Fuel elements were next arranged in a cylindrical assembly, products being fed through the core of a system of horizontally positioned spent fuel elements. Such a device was operated in air, and is shown in Fig. 109. It was possible to have a continuous process through such a unit, indicated in the figure.

At the Savannah River Food Irradiation Facility, using selected spent fuel elements a few days after being removed from a reactor, it has been possible with a suitable array of fuel elements to sterilize a 6-inch diameter by 7-inch long package of roast beef, a ham, or turkey with ionizing radiations in a matter of 20 minutes. A steady stream of such packages can be processed. A half carcass of beef in a suitable container could be sterilized in a similar time, provided that the array of spent fuel elements of sufficient activity and required configuration is accumulated.

Such facilities are useful in developing products and processes, but yield little information of value in terms of the economic feasibility of such processes. A prototype production facility is required to establish the economics of the process. Operating with a facility designed for the process rather than with some makeshift arrangement will yield the first useful data on the economic aspects of radiation technology. In the absence of an actual large scale irradiation facility, the problem of estimating irradiation costs is exceedingly difficult. To date, no completely satisfactory information exists.

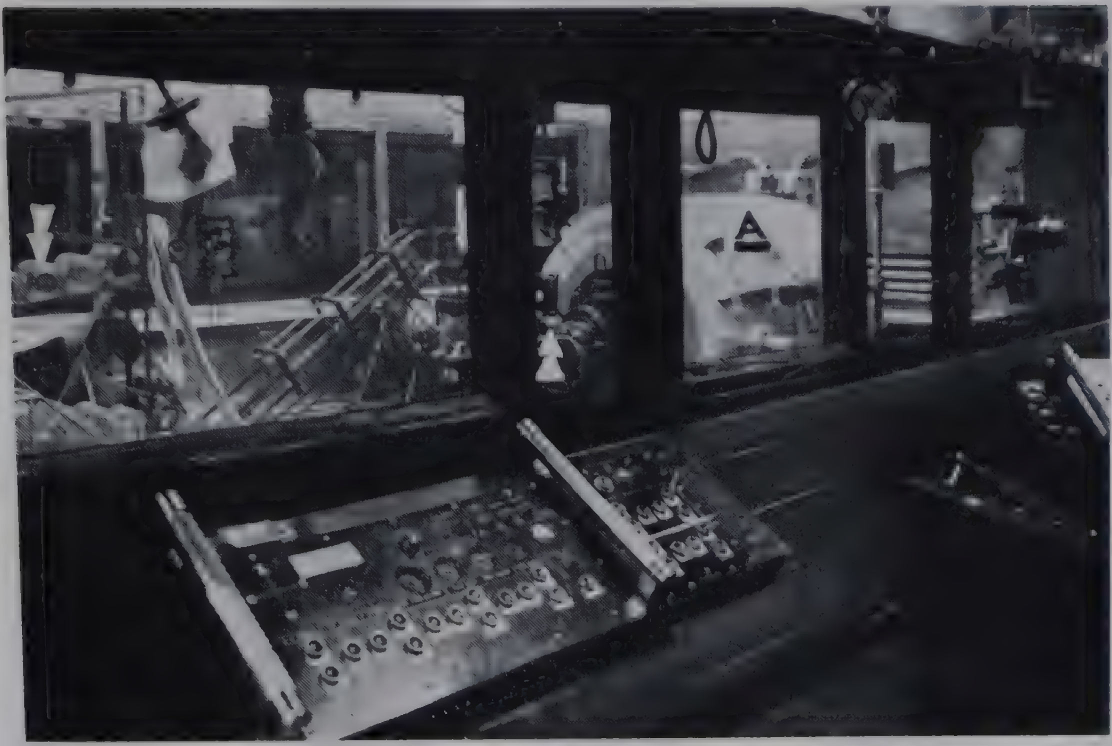


Courtesy of Phillips Petroleum Co.

Fig. 108. Early Radiation Facility Using Spent Fuel Elements at Idaho Falls, Idaho

Design of Radiation Processing Food Plant

The U.S.A., U.K., France and the U.S.S.R. have designed food processing plants which use radiation as the sterilizing agent.



Courtesy of U.S. Army Quartermaster Corps

FIG. 109. HIGH INTENSITY GAMMA RADIATION FACILITY AT DUGWAY PROVING GROUNDS, DUGWAY, UTAH

A radiation processing plant differs little from a conventional canning or freezing plant, with the exception of the preservation areas. A diagram of a mobile irradiation process is shown in Fig. 110.

A series of design concepts are shown in Figs. 111, 112, 113, and 114 to demonstrate the wide range of possibilities available in the development of a useful radiation technology. Other electron generating devices were shown in Figs. 20, 21, 23, and 24 (Chapter 6).

Radiation Source and Process Economics

Radiation technology is at such an early stage of development that the process does not lend itself to the usual cost analyses that precede the installation of new products and new processes into an industry. But with the fact in mind that radiation will grow into a major factor in the economy, it is necessary to make some evaluation of radiation cost factors, at least to form a basis for determining the pathway along which radiation technology will develop. This area of activity has been sponsored by the Office of Isotope Development of the U.S. Atomic Energy Commission, the information below being abstracted from such research reports Johnston 1959, Little 1959).

Relation between process capacity and source strength

The quantity of product per year, P, which is obtainable in a given radiation process may be defined by the following equation:

$$P = 0.0212\,G\,M\,C\,t\,f$$

where $P =$ the capacity in tons of product per year

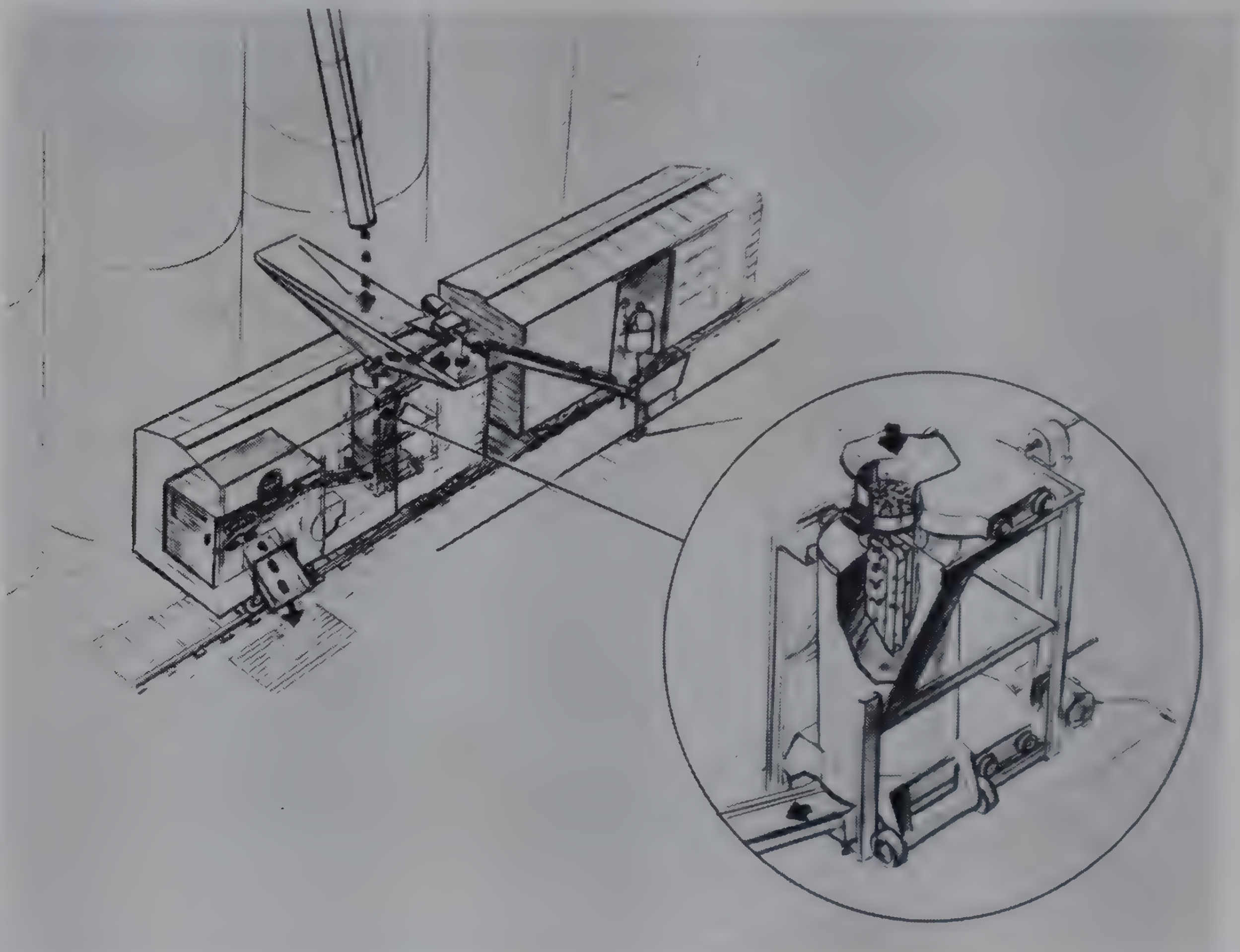
$G =$ the "G value" or energy utilization factor which is defined as the number of molecules of product generated per 100 electron volts of radiation energy absorbed.

$M =$ the molecular weight of the product

$C =$ the source strength in megacuries

$t =$ the energy per disintegration in Mev of the source material

$f =$ the geometric design efficiency factor (always less than unity) i.e., the fraction of the available radiation which is actually absorbed by the material being treated



Courtesy of U.S. Army Quartermaster Corps

FIG. 110. MOBILE IRRADIATION PROCESSING PLANT

Cobalt-60 is one of the most suitable of the materials used as radiation sources. For Co^{60}, one kw. = 0.084 megacuries (see Table 67). Hence, for an irradiator using Co^{60}, one megacurie = 11.90 kw., and the equation may be written

$$P = 0.001782\,G\,M\,K\,t\,f$$

where K = the effective source strength in kw. for Co^{60}.

TABLE 67

CONVERSION FACTORS

1 megarad = 0.00126 kw. hr.
1 kw. = 0.084 megacuries Co^{60}
1 kw. = 0.402 megacuries Cs^{137}
1 kw. = 0.303 megacuries Zr-Nb

The quantity K will depend on the quantity of Co^{60} in the particular irradiator under consideration and is a measure or rating of the size or capacity of the facility.

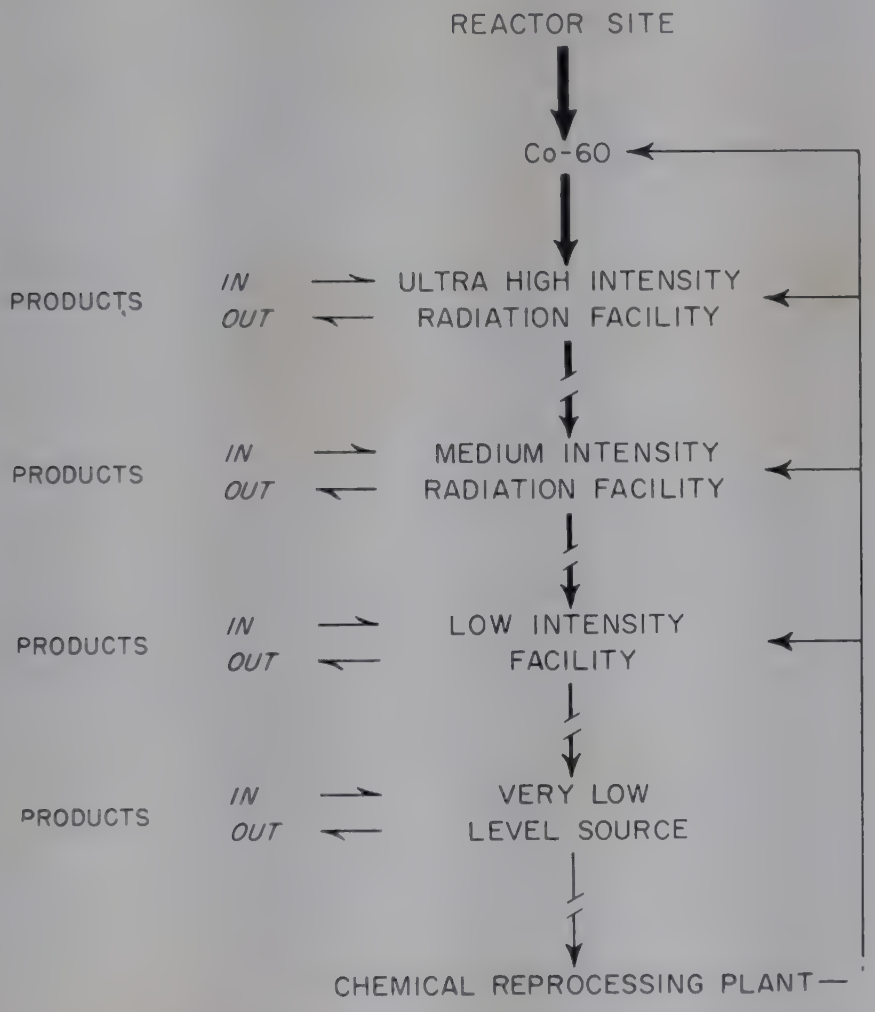

REACTOR SITE
Co-60
PRODUCTS
IN
OUT
ULTRA HIGH INTENSITY RADIATION FACILITY
PRODUCTS
IN
OUT
MEDIUM INTENSITY RADIATION FACILITY
PRODUCTS
IN
OUT
LOW INTENSITY FACILITY
PRODUCTS
IN
OUT
VERY LOW LEVEL SOURCE
CHEMICAL REPROCESSING PLANT

FIG. 111. HIGH SPECIFIC ACTIVITY RADIONUCLEIDE FACILITIES WILL "FEED" LOWER INTENSITY FACILITIES, IMPROVING COSTS OF RADIATION PROCESSING

The higher volume higher specific activity sources obtain radiation at lower cost per curie than low intensity sources. Cost per curie is less for 10,000,000 curies than for 10 curies. Each facility operates on a "radiation used" basis. Radioisotopes will not be discarded in the future if useful.

For Co^{60}, $t = 2.5$ Mev (an inherent property of the isotope) hence, the equation becomes

$$P = 0.004455\, G\, M\, K\, f$$

To illustrate the application of this equation, consider a Co^{60} irradiator of moderate size rated at 30 kw. maintained at reasonably constant power level by isotope replacement. It is desired to promote the production of phenol from gaseous benzene-oxygen mixtures. For this operation,

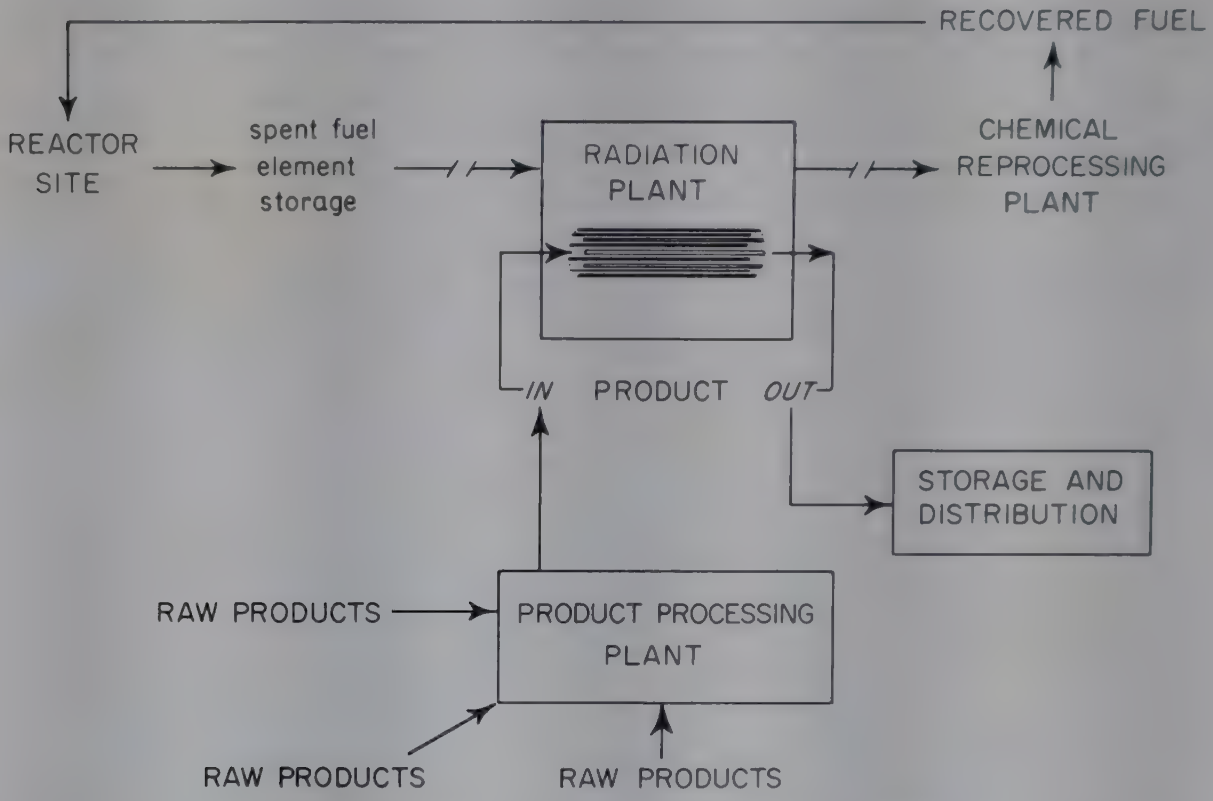

RECOVERED FUEL
REACTOR SITE
spent fuel element storage
RADIATION PLANT
CHEMICAL REPROCESSING PLANT
IN
PRODUCT
OUT
STORAGE AND DISTRIBUTION
RAW PRODUCTS
PRODUCT PROCESSING PLANT
RAW PRODUCTS
RAW PRODUCTS

FIG. 112. SPENT FUEL ELEMENT RADIATION FACILITY—INTERCEPTION OF FUEL ELEMENTS BETWEEN REACTOR AND CHEMICAL REPROCESSING PLANT

$G = 214$ (determined by previous experiment)
$M = 94$ for phenol
$K = 30$ kw.
$f = 0.8$ (assumed for this irradiator)

hence $P = 0.004455 \times 214 \times 94 \times 30 \times 0.8$
$= 2151$ tons of phenol per year

The equations for the cobalt-60 sources are applicable to machine or other sources as well. The choice of a source will depend on its configuration, method of employment, efficiency, and relative cost. The production of useful materials using high level radiations is often possible, but the cost factors of both process and the sources must be discussed, to have any meaning.

Radiation Economics

The cost of radiation in terms of dollars per kilowatt hour consumed forms a basis for measuring radiation production costs. Those products with high *G* values and high molecular weights have low radiation processing costs per pound. The United States Atomic Energy Commission has conducted numerous studies on the economics of industrial radiation processing. The results of such analyses for different radiation systems at both a 3 and a 30 kilowatt level have been summarized and are presented in Tables 68, 69, 70, and 71. There are four considerations: (1) source size and machine size, (2) nuclide costs vs. machine costs, (3) operation and maintenance costs, and (4) cost analysis. The cost per kilowatt hour for radiation can be subdivided into four large categories: the cost of the

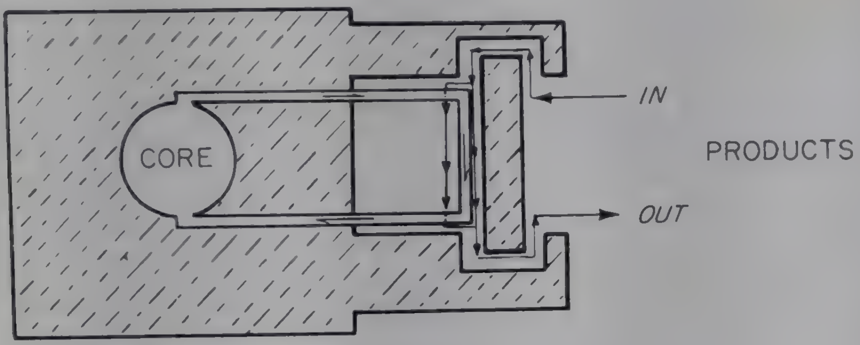


FIG. 113. REACTOR CORE LOOP—A PERMANENT RADIATION SITE

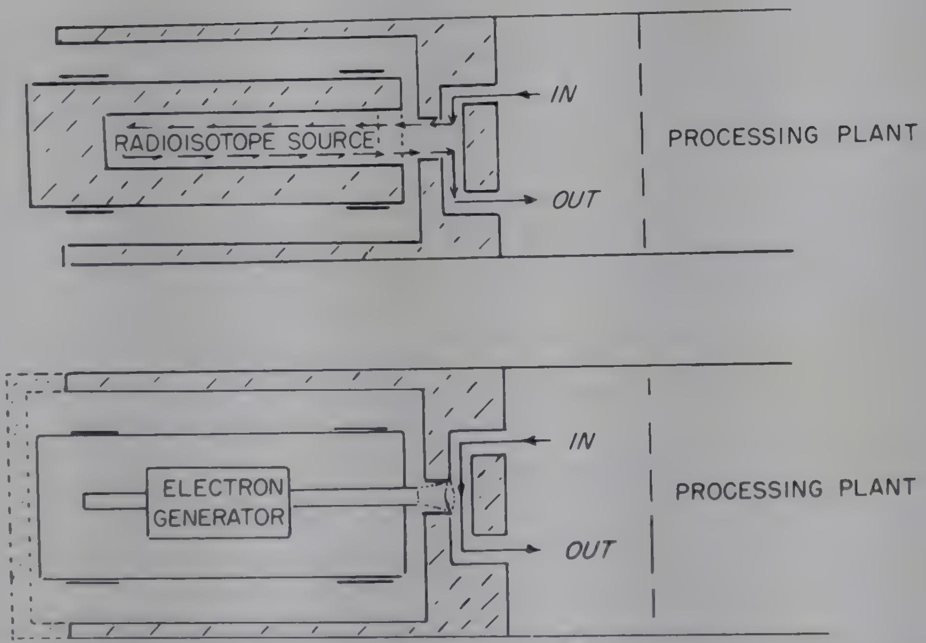


FIG. 114. MOBILE RADIATION SOURCE. RADIONUCLEIDES OR ELECTRON GENERATORS CONTAINED IN MOBILE BEDS OR VANS. SHIELDING REQUIRED AS INTEGRAL PARTS OF THE STRUCTURES

TABLE 68

SUMMARY COST ANALYSIS OF MACHINE AND GAMMA NUCLIDE SOURCES[1]

Item	Machine 3 kw.	Cobalt-60 3 kw.	Cesium-137 3 kw.	Zirconium Niobium 3 kw.	Machine 30 kw. 2–15 kw. Mach.	Cobalt-60 30 kw.	Cesium-137 30 kw.	Zirconium Niobium 30 kw.
Purchase price of machine or source preparation	$ 66,000	$159,000 12,600	$304,000 121,600	$ 36,300	$300,000	$1,260,000 126,000	$1,825,000 1,216,000	$181,600
Shipment (1,000 miles)	500	2,500	12,200		4,500	25,000	121,600	
Installation	45,000	120,000	120,000	120,000	90,000	300,000	300,000	300,000
Total capital investment	111,500	324,100	557,800	156,300	394,500	1,711,000	3,462,600	481,600
Depreciation (10 year, Straight-line)	11,150/yr.	32,400/yr.	55,800/yr.	15,600/yr.	39,450/yr.	171,100/yr.	346,300/yr.	48,200/yr.
Operation and maintenance	138,000/yr.	125,900/yr.	111,500/yr.	222,000/yr.	265,500/yr.	278,800/yr.	171,300/yr.	702,800/yr.
Total annual cost *excluding* return on investment	149,150/yr.	158,300/yr.	167,300/yr.	237,600/yr.	304,950/yr.	449,900/yr.	317,600/yr.	751,000/yr.
Cost per kw. hr. produced excluding return on investment	6.95/kw. hr.	6.60/kw. hr.	7.00/kw. hr.	10.50/kw. hr.	1.40/kw. hr.	1.90/kw. hr.	2.15/kw. hr.	3.30/kw. hr.
Provision for return on investment (30% of average investment)	16,725/yr.	48,600/yr.	83,700/yr.	23,400/yr.	59,175/yr.	256,700/yr.	519,300/yr.	72,200/yr.
Total annual cost *including* return on investment	165,875/yr.	206,900/yr.	251,000/yr.	261,000/yr.	364,125/yr.	706,600/yr.	1,036,900/yr.	823,200/yr.
Cost per kw. hr. produced *including* return on investment[2]	7.75/kw. hr.	8.70/kw. hr.	10.50/kw. hr.	11.50/kw. hr.	1.70/kw. hr.	2.95/kw. hr.	4.35/kw. hr.	3.65/kw. hr.

---

[1] From Michaelis (1959).

[2] Based on an operating schedule of 8,400 hours per year, less 15% allowance for downtime.

Table 69

MACHINE AND NUCLIDE SOURCE COSTS[1]

Machine Rating			Purchase Price, $/kw.
Kw.	Mev		
3	1.5–3.0		$22,000
30	1.5–3.0		$10,000
Nuclide Sources	Nuclide Price, $/curie		
Source size	Cobalt 60	Cesium 137	Zr-Nb
3 kw.	$0.75	$0.25	$0.04
30 kw.	$0.50	$0.15	$0.02
	Curie Requirements[2]		
3 kw.	252,000	1,216,000	908,000
30 kw.	2,520,000	12,160,000	9,080,000

[1] From Michaelis (1959).

[2] These source requirements allow for reasonable self-absorption losses and overdesign to give rated capacity at end of replacement period.

radiation source, the cost of the supporting facilities, the cost of operating manpower, and the maintenance and replacement costs.

From these tables (68–71) it is seen that machine radiation costs per kilowatt hour can be approximated by the equation:

Table 70

SUMMARY COST ANALYSIS OF BETA-NUCLIDE SOURCES[1]
(30 kilowatt power level)

	Ce^{144}	Pn^{144}	Ru^{106}	Zr^{90} (fluoride)	Zr^{90} (oxide)
Purchase price of source	2,034,000	28.800,000	1,680,000	2,184,000	2,184,000
Source fabrication	120,750	1,068,000	47,700	163,800	411,000
Total capital investment†	2,154,750	29,868,000	1,717,700	2,347,800	2,595,000
Depreciation (10 yr. st. line)	215,475	2,986,800	172,770	234,780	259,500
Replacement costs††	1,736,400	7,314,000	1,134,000	55,200	60,810
	1,951,875	10,300,800	1,206,700	289,980	320,311
$/kw. hr.†††	$8.94	48.10	6.10	1.35	1.50

† Not including installation and shipping.
†† Not including personnel salaries and overhead.
††† Based on 8,400 hrs./yr. less 15 per cent allowance for downtime. Not including return on investment.
From Johnston (1959).

$$\frac{\text{Dollars}}{\text{kilowatt hour}} = \frac{21.18}{K \times 0.66}$$

where K is the power level of the machine installation in kilowatts.

According to one estimate, shown in Table 68 the cost per kilowatt hour for a 30 kilowatt installation ranges from $1.70 to $4.35, depending

TABLE 71

COMPOSITION OF Co^{60} RADIATION PRODUCTION COSTS FOR DIFFERENT SOURCE POWER LEVELS[1]

Source Power Level (Kilowatts)	Labor and Overhead Basic Parts and Utilities Basic Capital Investments	Capital Investment and Parts and Utilities which are Proportional tc Plant Size	Total Isotope Costs Including Replacements, Source Preparation, and Shipping	Total Cost of Radiation Production $/kw. hr.	Curie cost
3	$4.68	$0.083	$1.33	$6.08	$0.56
30	0.468	0.083	1.33	1.88	0.56
100	0.140	0.083	1.33	1.55	0.56
300	0.047	0.083	1.33	1.46	0.56
3	4.68	0.083	0.27	5.03	0.11
30	0.468	0.083	0.27	0.82	0.11
100	0.140	0.083	0.27	0.49	0.11
300	0.047	0.083	0.27	0.40	0.11

[1] From Johnston (1959).

on the source characteristics. If the installation is increased to 300 kilowatts and the number of operating personnel is the same, the cost could be substantially lower.

According to another study, the radiation cost for a radioisotope source, assuming that all capital costs are amortized in ten years, can be expressed as follows:

$$E = \frac{1}{f}\left(\frac{13.32}{K} + 11.91 + ND_c\,(1+10R) + 0.08\right)\left(\frac{1}{1-n}\right)$$

where E = the cost per kilowatt-hour in dollars
N = the power equivalent in megacuries per kilowatt
D_c = the cost per curie including preparation and shipping charges
R = the fraction of the radioisotope replaced annually in order to maintain the desired power level
n = the fraction of time during which the source is not in use
f = an efficiency factor of utilization of radiation
K = effective source strength in kilowatts

The first term, $\frac{13.32}{K}$, in the equation indicates the part of the radiation cost which is borne by labor and general overhead. These are relatively independent of the source size within the range of 300 kw. In effect, to increase the power level of the radiation facilities lowers unit cost, indicating the trend and advantage in operating a large rather than a small facility. These costs can be reduced by amortizing for longer periods of time or by reducing the labor costs for operating the facility. The latter may follow directly from larger scale facilities. At present it is feasible to build 30 kilowatt sources and 300 kilowatt sources appear to be not out of reach. It appears that the most important factor in determining the cost per kilowatt hour is the isotope cost. Typical values calculated from the above equation are shown in Table 71.

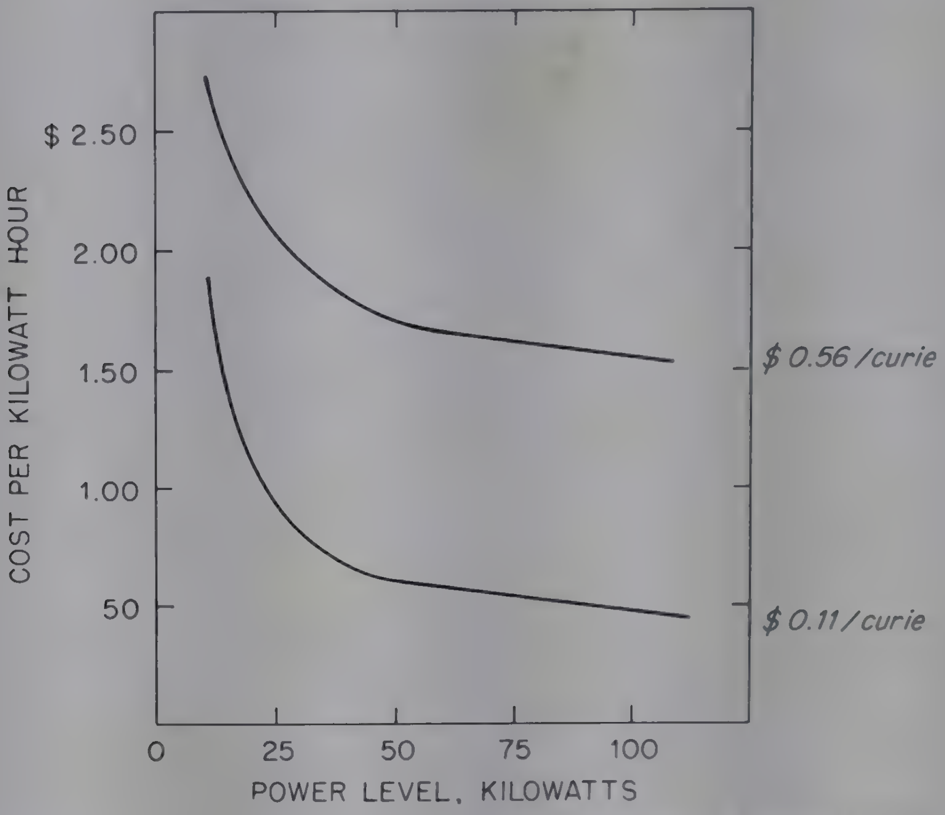
Cost on a Radiation "Used" Basis

Since the source level is maintained by periodic replacement of the source elements, at the end of a given time, the source still contains essentially the same activity it had at the beginning. Therefore, it is actually unreasonable to amortize the full initial cost of the isotopes over any particular time. It appears more reasonable to consider solely the cost of using the emitted radiation rather than the entire cost of the isotope itself. The problem is in some respects similar to the cost accounting of a platinum catalyst. After the process has terminated, the platinum is largely recoverable and has value. Similarly, the radioisotope is still useful although it may now be of lower strength. In order to maintain a source at a given level, it is assumed that 12 per cent of the isotope is replaced annually. The source elements must be replaced when they have decayed approximately 12 per cent. Such replaced elements are still of very high activity. It would appear that as much as 80 per cent of the investment must be considered as a recoverable quantity, provided no drastic isotope price changes occur over these eight years. If the price does decrease, the recoverable cost decreases, but so does the replacement cost.

It appears that some part of the problem of projected high radiation costs must be attributed to the preliminary nature of cost assessments in the absence of operational data. The real cost of the nuclide should be actually the amount of decrease in activity experienced during its use. It would appear that what is required is some mechanism for supply and replacement of radiation source nuclides to industry. Some form of co-operative effort is indicated between government and industry in this regard. From Fig. 115 it is seen that no matter what the price of a curie, large power sources are more economical than small. This is but an example of economy of scale found in many processing systems.

An enormous amount of high level radiation will be required for the successful application of ionizing radiation solely in food stabilization. The amount of radiation required on the average to sterilize a pound of food (meat) is 4.8 megarads which corresponds to 0.0063 kilowatt hours per pound. In a 300 kw cobalt-60 radiation source, the cost per kilowatt hour for radiation is $1.46 for 56 cent cobalt-60 or 40¢ for 11 cent cobalt-60 curies. Since cobalt-60 for this purpose may be considered a by-product, its price will be subject to fluctuation depending on the use to which nuclear energy is put. For these cases, radiation sterilization costs at 5 megarad dose levels would be between 0.92 and 0.25 cents per pound respectively (Table 71). Even at the higher cost, 0.92 cents, it is still competitive with steam sterilization at current costs.

When we consider the annual world market for food, it amounts to some 4 × 10^{13} pounds, i.e., 40,000,000,000,000 pounds per year. Suppose we assume that sterilization is useful for only one per cent of this food, that is, on 400,000,000,000 pounds per year. There will be an annual requirement for this purpose alone of about 2.5 billion kilowatt hours. Using cobalt-60 as an example, this would be approximately 212,000,000,000 megacuries of cobalt-60 at an annual replacement of 12.5 per cent to maintain the source level at the desired amount. It would appear that with just food irradiation alone, the future is relatively secure for suppliers of all forms of radiation (Johnston *et al.* 1959).



Courtesy of Johnston (1959)

FIG. 115. RELATION OF COST TO SIZE IN AN IRRADIATION FACILITY

While only fragmentary information is available pertaining to reactor costs as a primary source of radiation, the few cost analyses which have been made are moderately encouraging. One oversimplification has been the assumption that a single product would have to bear the whole cost of the installation. It would seem more reasonable to use reactors for multiple purposes including power production and radiation applications. Careful analysis is indicated.

In summary of the economics of radiation processing, it would appear that, as larger irradiation facilities are built, the unit cost of processing will go down. When the cost of radiation is computed on the basis of actual curies consumed rather than the cost of producing the curie, the radiation costs are sufficiently low, even perhaps for machine-generated radiation sources, to yield processes of substantial financial appeal and value. Obviously any actual cost basis for programming radiation source costs will require extensive study into isotope economics, supply, demand, and resale. Tables 68–71 give a summary of some cost analyses of the machine and radioisotope sources, sponsored by the United States Atomic Energy Commission. These do not include the consideration of radiation costs based on the curies used basis, which will appear soon.

In spite of the fact that radiation processing has not developed to the point where completely realistic cost analyses can be made, with the information available relative to the "curies-used" concept, it would appear that radiation installations could *now* be undertaken yielding product irradiations either equal to or lower in cost than conventional steam sterilization, for example.

There are many avenues of approach in evaluating radiation processing and its economics. If a spent fuel element removed from power reactors is in transit to a chemical reprocessing plant, it would appear likely that these radiation sources could be intercepted at some convenient point and be usefully applied. In such an event, a food processor or drug manufacturer might well consider the routing of a product from the factory through an adjacent radiation device, back to the plant. The intercepted fuel elements could be a mobile radiation chamber and considered on the other hand as a component of the factory source. The utilization of such devices as the sodium loop from a power reactor for gamma irradiation of useful quantities and qualities, will certainly warrant favorable economic consideration. While such radiation sources currently are not programmed (that is, the cost of power production does not depend on the ability to use gamma irradiation usefully) certainly it may be envisioned that the cost for radiation service could be one reasonably and usefully priced.

It is apparent that the cost per kilowatt hour of high energy radiation may, for some time to come, be considerably higher than the cost of a kilowatt hour of conventional energy obtained from fossil fuels or hydroelectric facilities. These two types of power must under no circumstances be considered as equivalent with regard to the effect that they can induce in materials. The amount of conventional energy, e.g., through the medium of steam, that must be imparted to a pound of beef to sterilize it is much greater than the amount of radiation energy needed to accomplish the same ends. The unique characteristic of high energy radiation, the theme of this book, is that radiation can produce major effects with comparatively small energy inputs.

While the economics of radiation technology are still at an early stage of development, it appears that the process is useful, is practical and favorable costwise with presently available systems of performing the same task. Realizing the tremendous impact the economics of food distribution has on the health and welfare of mankind, the opportunity of usefully applying radiation appears boundless and economically favorable.

BIBLIOGRAPHY

Aebersold, P. C. 1955. Importance of isotopes in technology and industry. Proc. Intern. Conf. Peaceful Uses of Atomic Energy *14*, 3–20. Geneva.

Aebersold, P. C. 1959. The United States industrial radiation program. Intern. Atomic Energy Agency, Warsaw Conference. Paper CW/S/57.

Artandi, C., and Van Winkle, W., Jr. 1959. Electron beam sterilization of surgical sutures. Nucleonics *17*, No. 3, 86–88.

Atomic Industrial Forum, Inc. 1955. A growth survey on the atomic industry, 1955–1965. New York, N. Y.

Brynjolfsson, A., and Holm, N. W. 1959. The cobalt-60 irradiation facility and gamma field at Rise, Denmark. International Atomic Energy Agency Warsaw Conference. Paper CW/Ipl/75.

Burrill, E. A., and Gale, A. J. 1952. Electron beams sterilize food and drugs. Electronics *25*, 98–101.

Christensen, C. J., Jacobsen, A., Klinger, Q., and Abel, G. 1957. Dugway food irradiator uses 8 MTR fuel elements. Nucleonics *15*, No. 12, 94–96.

Colichman, E. J., Mallon, P. J., and Jarrett, A. A. 1957. Gamma facility for use at high temperatures. Nucleonics *15*, No. 4, 115–118.

Desrosier, N. W. 1958. Food irradiation research and its practical application in Europe and the United States. Project Rept. No. 396. E.P.A.-O.E.E.C. 2 Rue Andre Pascal, Paris, France.

Desrosier, N. W. 1959. The Technology of Food Preservation. Avi Publ. Co., Inc., Westport, Conn.

Fahnoe, F., Crean, L., and Kupp, R. 1952. Industrial applications of gross fission products. U. S. Atomic Energy Commission Con. No. AT(30-1)-850. Germantown, Maryland.

George, D. W., and Gregory, J. N. 1959. Gamma radiation facilities at the Australian Atomic Energy Commission Research Establishment. Intern. Atomic Energy Agency, Warsaw Conference. Paper CW/Ipl/12.

Glasstone, S. 1958. Sourcebook on Atomic Energy. Second Ed. D. Van Nostrand and Company, New York, N. Y.

Hannan, R. S. 1956. Science and Technology of Food Preservation by Ionizing Radiations. Chemical Publ. Co., New York, N. Y.

Harlan, J. T., and Hart, E. J. 1959. Ceric dosimetry: accurate measurements at 10^8 rads. Nucleonics *17*, No. 8, 102–104.

HARTECK, P., and DONDES, J. 1956. Producing chemicals with reactor irradiation. Nucleonics *14*, No. 7, 22–25.

HARTECK, P., and DONDES, S. 1957. Glass fibers—a new form for reactor fuels. Nucleonics *15*, No. 8, 94–98.

HUBER, W. 1948. Sterilization of drugs. Electronics *21*, 74–79.

INTERN. ATOMIC ENERGY AGENCY. 1959. Abstracts of papers for the conference on the application of large radiation sources in industry. Vienna, Austria.

JEFFERSON, S., MURRAY, G. S., and ROGERS, F. 1959. Gamma radiation sources of the Great Britain Technological Irradiation Group. Intern. Atomic Energy Agency, Warsaw Conference. Paper CW/Ipl/87.

JOHNSTON, W. H., WALLENSTEIN, M. B., ROSENSTOCK, H. R., and DESROSIER, N. W. 1959. Industrial Role of Radiation. Research Rept. Con. AT(11-1)-650. U. S. Atomic Energy Commission, Office of Isotope Development, Germantown, Maryland.

KUHL, O. A., SPARROW, A. H., and MANOWITZ, B. 1955. Portable pilot plant for irradiating potatoes. Nucleonics *13*, No. 11, 128–131.

LEVEQUE, P. 1959. Description of sources for irradiation (in French). Intern. Atomic Energy Agency, Warsaw Conference. Paper CW/Ipl/40.

LIBBY, W. F. 1957. Radioisotopes. Nucleonics *15*, No. 9, 118–121.

LITTLE, A. D., INC. 1959. Radiation: A Tool for Industry. Con. Rept. Office of Isotope Development, U. S. Atomic Energy Commission, Germantown, Maryland.

MANOWITZ, B., KUHL, O. A., FALANTER, L., BRETTON, R. H., and SWICKLER, S. 1959. Experimental parametric study of large scale cobalt, sodium, and cesium slab irradiators. Intern. Atomic Energy Agency, Warsaw Conference. Paper CW/Ipl/75.

MANOWITZ, B., and RICHMAN, D. M. 1956. Economic future of fission products for radiation power. Nucleonics *14*, No. 6, 96–101.

MATSUMURA, S., FUJII, T., and KONDO, S. 1959. Cobalt-60 gamma irradiator and its uses in plant genetics in Japan. Intern. Atomic Energy Agency, Warsaw Conference. Paper CW/IVA/30.

MICHAELIS, M. 1959. Technology and economics of large radiation sources. Intern. Atomic Energy Agency, Warsaw Conference. Paper CW/IIC/19.

NUCLEONICS REPORT. 1958. Understanding nuclear power costs. Nucleonics *16*, No. 1, 46–57.

NUCLEONICS REPORT. 1959. Food irradiation looks to low dose treatments. Nucleonics *17*, No. 10, 112–113.

POMERANTZ, P. and SIU, R. G. H. 1957. Economics of radiation processing Chap. 22. Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

Progress Report on Atomic Energy Research. 1956. Hearings before the Committee on Research and Development of the Joint Committee on Atomic Energy, Congress of the U. S., 4–8 June. U. S. Govt. Printing Office, Washington, D. C.

RANFTL, J. W. 1959. Irradiated polyethylene—a case study in economics of production uses of electron beam radiation. Intern. Atomic Energy Agency, Warsaw Conference. Paper CW/IIIp/54.

RICE, F. G., COURTEMANCHE, R., and MASEFIELD, J. 1959. Preliminary design study of a mobile irradiator. Intern. Atomic Energy Agency, Warsaw Conference. Paper CW/I/20.

SILOW, R. A. 1958. The potential contribution of atomic energy to development in agriculture and related industries. Intern. J. Applied Radiation and Isotopes *3*, 257–280.

STANFORD RESEARCH INSTITUTE. 1951. Industrial uses of radioactive fission products. Report to the U. S. Atomic Energy Commission, Project 361.

SUN, K. H. 1955. A comparison of radiation sources and some typical applications in the chemical industry. Atomic Energy in Industry Conference. New York, N. Y. Oct. 26–28.

SYMPOSIUM ON THE INDUSTRIAL ROLE OF RADIATION. 1959. Georgia Institute of Technology, Atlanta, Georgia.

THOMAS, A. 1955. Nuclear batteries: types and possible uses. Nucleonics *13*, No. 11, 129–132.

UNITED NATIONS. 1958. Peaceful Uses of Atomic Energy. Proc. of the Second Intern. Conference, Geneva. 33 volumes.

U. S. ARMY QM. CORPS. 1957. Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

UNITED STATES JOINT COMMITTEE ON ATOMIC ENERGY. 1956. Peaceful Uses of Atomic Energy. Report of the Panel on the Peaceful Uses of Atomic Energy. U. S. Govt. Printing Office, Washington, D. C.

WALLENSTEIN, M. B., ROSENSTOCK, H. M., and JOHNSTON, W. H. 1959. Fundamental Considerations of Radiation Technology. U.S.A.E.C. Con. Rept. AT(11-1)-650. Parts I, II.

CHAPTER 21

Radiological Safety

A radiological safety program is of primary importance in any installation using sources of ionizing radiation. The safety program for a radiation facility is highly involved, and health physics is a field in itself. The sources employed in radiation technology will be a radioisotope, a reactor, or an accelerator. While the discoverers of radioactivity and many radiologists working in the early parts of this century were injured due to their association with radioactive substances, at the present time safety methods have been developed to the point where personnel are as safe in such a facility as in any other plant. The safety record of the Oak Ridge National Laboratory is an example (Morgan 1947). However, only by having a working knowledge of the hazards and by constantly applying the proper techniques can the desired safety be insured to personnel. It is useful to review briefly human radiation tolerances, surveying and monitoring systems, radiation shielding, Federal regulations governing the operation of radiation facilities, and the problem of contamination of land masses due to accidents.

Human Radiation Tolerances

As has been discussed in Chapter 14, whole body radiation has early, late, and delayed effects. The physiological effects on a human being due to the absorption of an acute dose of gamma radiation distributed over the entire body are summarized in Tables 72 and 73.

It should be noted that there is some uncertainty concerning the absolute effects of a given dose of radiation to man. Part of this is due to the lack of data relative to mankind, and part is due to biological variations which are commonly encountered (Cooper and Siu 1957). The clinical symptoms following radiation exposures manifested in radiation sickness are listed in Table 59. The various parts of the human body vary substantially in their response to radiation. The nuclei of cells appear to be the most sensitive (Claus 1958). However, those cells which are growing or reproducing rapidly are the most sensitive of all to damage. Examples are found in the lymphatic tissue and the bone marrow. In order of decreasing radiation sensitivity, the cells of the human body are arranged in the sequence of reproductive tissue and skin, blood vessels, connective tissue cells, muscle and bone cells, and finally nerve cells. The hazard due to the ingestion of radioisotopes inside the body is of great significance because of the long biological half-life of certain radioisotopes and their

TABLE 72

RADIATION EFFECTS DUE TO WHOLE BODY EXPOSURE IN A PERIOD OF ONE DAY OR LESS[1]

Dose (Rads)	Effects
0–25	No obvious injury
25–50	Possible reduction in white blood cell count, no permanent disability
50–100	Nausea, fatigue, reduction in white blood cell count, recovery
100–200	Reduction in white blood cell count, sore throat, vomiting, diarrhea, recovery possible with some disability
200–400	Permanent disability, death possible
400	Fatal to half of population, permanent disability in survivors
600–	Fatal to all population

[1] From Cooper and Siu (1957).

intimate contact with body substances. And, local concentration of particular isotopes such as strontium-90 or iodine-131, may become excessively high due to preferential localization of the elements in particular parts of the body.

Mankind lives in a constant low level field of radiation, as discussed in Chapter 1. The radiation which man can absorb during a 30-year period of life on earth is summarized in Table 74. At the present time, there is great controversy over the effects of radiation, and these discussions center around the rate of production of mutants and the significance of these mutants in future generations of mankind. It is to be noted from Table 74 that the artificial sources of radiation produce an exposure in man

TABLE 73

CLINICAL SYMPTOMS OF RADIATION SICKNESS[1]

Time Following Exposure	Dose Received (Rads)		
	600	400	100–300
7 days	Nausea after 1–2 hrs. Reduction in white blood cell count Fever Inflammation of mouth and throat	Nausea after 1–2 hrs. Reduction in white blood cell count No definite symptoms otherwise	No definite symptoms
14 days	Rapid emaciation Death	Low white blood cell counts Loss of appetite Fever	No definite symptoms Lowered white blood cell counts
21 days	—	Fever Inflamed mouth and throat Lower white blood cell count	Loss of appetite Sore throat Diarrhea
28 days	—	Rapid emaciation 50 per cent of population dead Others permanently injured	Diarrhea Moderate emaciation Recovery likely Some permanent injury

[1] From Cooper and Siu (1957).

which is roughly equal to that from unavoidable natural background radioactivity which he cannot escape. Because of the potential danger to future generations of mankind, a number of governmental agencies have developed codes for radiological safety for the guidance of people working in this area. These include the International Commission on Radiological Protection established in 1928 under the sponsorship of The International Conference of Radiobiology. The United States Committee on Radiation Protection was established in 1928, and the National Bureau of Standards in the U. S. A. issues its recommendations in the form of handbooks, Handbook 52 (U. S. Bureau of Standards 1953) being cited in reference most widely. The recommendations of various responsible agencies may be summarized as follows: For human adults under 45 years of age, the maximum permissible dose per week is 0.3 rads to the blood forming organs, the gonads, and the lenses of the eyes, and a weekly permissible dose of 0.6 rads to the skin. For personnel 45 years of age or older the weekly permissible doses are considered safe at double the above rates. However, for adults of any age, the maximum permissible exposure to radiations of low penetrating power is 1.5 rads per week, provided that the dose to the lens of the eye is less than 0.3 rads. The above recommendations apply to radiations below 3 Mev, and for personnel under surveillance.

TABLE 74

RADIATION RECEIVED BY HUMAN AFTER 30 YEARS ON EARTH UNDER ORDINARY CIRCUMSTANCES[1]

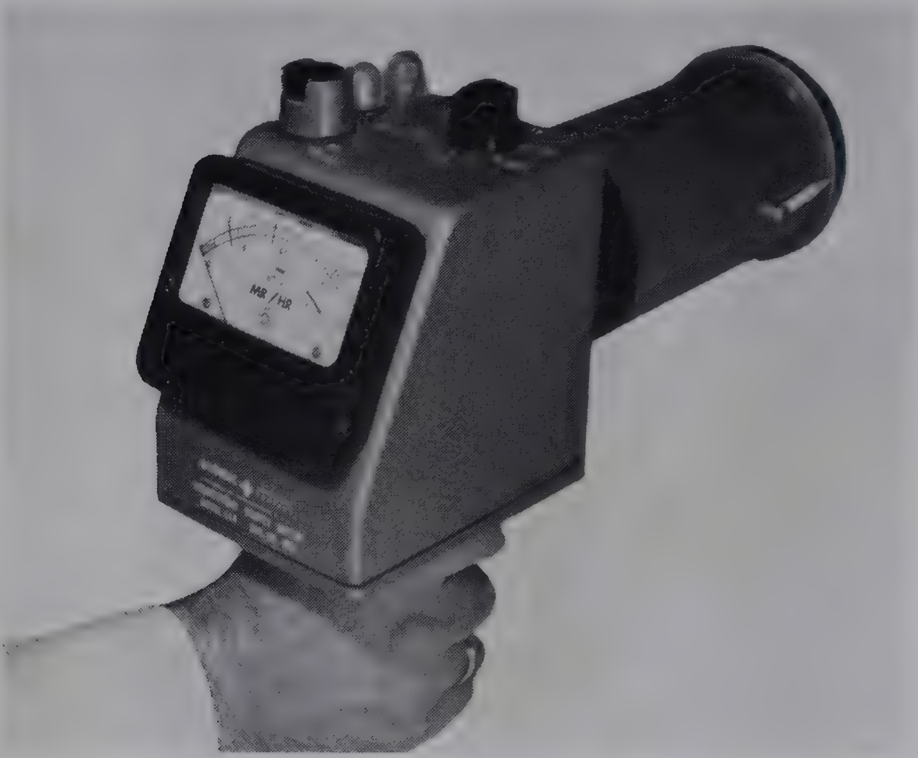
Source	Accumulated Dose in 30 Years (Rads)	Per Cent of Total
Natural background		
cosmic rays	1.1	15
earth	2.4	32
internal from ingested radioisotopes	0.8	11
Artificial sources		
medical examinations	3.0	40.5
wrist watches (luminous dials)	0.03	0.4
radioactive fallout	0.085	1.1
Total	7.415	100

[1] From Nat. Acad. Sci. (1956).

The adult may recover with no permanent effects from a whole body irradiation dose of 25 rads or an exposure to the hands or the feet up to 100 rads. Women in the reproductive stage should not be subjected to deliberate exposures of the above doses unless there is some other (medical) reason which makes it necessary. These regulations obviously are intended to be a guide, and compliance is essential for personnel safety. The commonly employed medical chest X-ray examinations each contribute approximately 0.6 rads. The use of X-ray techniques must obviously be restricted in use with humans. In the U. S. A., the National Academy of Science has recommended that the combined dose from all but normal background radiation for the first 30 years of life of a human being should be limited to 10 rads. It has also been recommended that radiation exposure records be maintained for the entire human population.

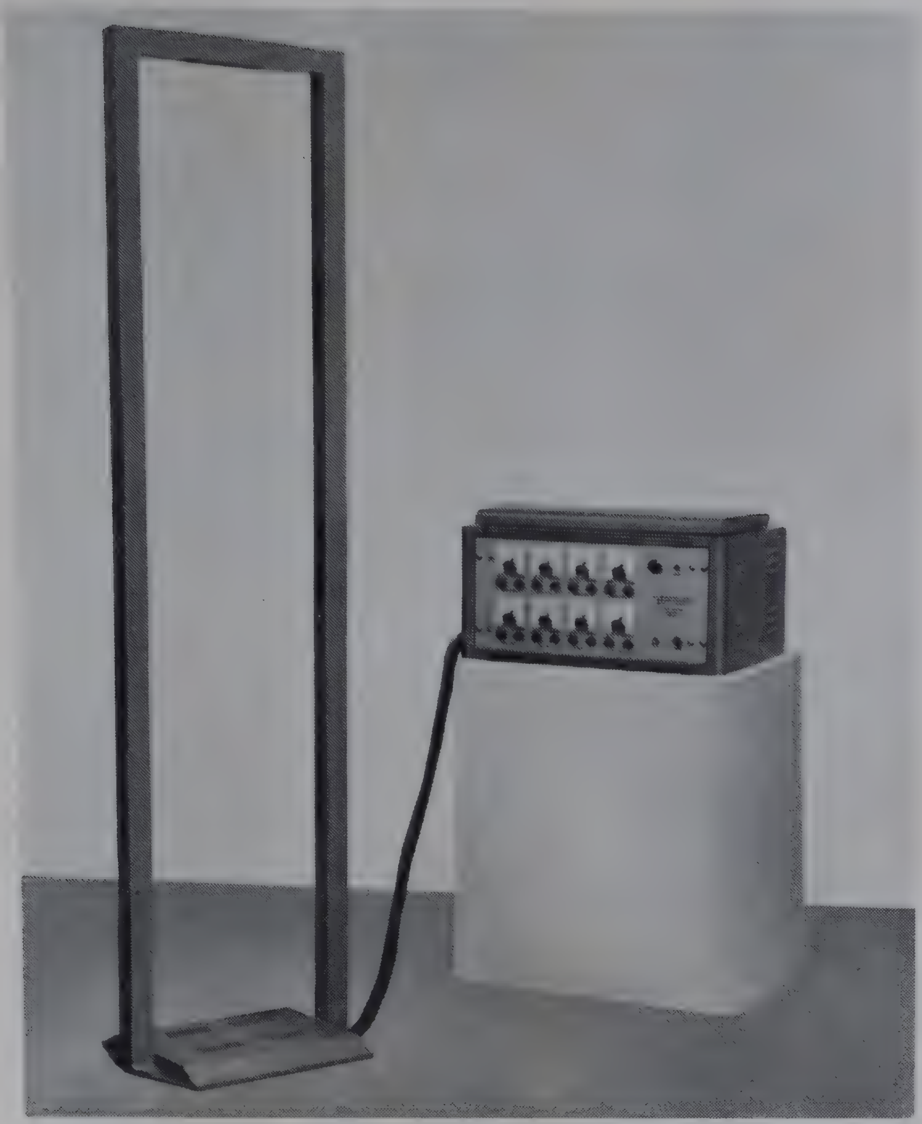
Surveying and Monitoring Systems

To establish safe working conditions for personnel, it is essential that all operating procedures be adequately planned and executed. It is essential to evaluate the techniques and facilities employed to minimize the exposure of personnel. Further, it is important to measure the cumulative exposure of an individual over a period of time such as a week, or the duration of a particular processing run. It is essential to detect radioactive contaminations on surfaces, in the air, and in all plant effluents and products. All of these aims require the proper selection of surveying and monitoring instruments, their satisfactory operation, interpretations of the results obtained, and maintenance of proper records. There is a large variety of devices and systems which are each suited to one of the many monitoring and surveying situations which are encountered in a large irradiation facility. These devices include such items as hand and foot monitors, film badges, pocket dosimeters, area survey meters, ionization



Courtesy of Baird Atomic

FIG. 116. "CUTIE PIE"—OR GUN TYPE SURVEY METER



Courtesy of Eberline Corp.

FIG. 117. PORTAL MONITOR—USEFUL IN DETECTING RADIOACTIVE CONTAMINATION ON PERSONNEL AS THEY PASS THROUGH AREA

chambers, atmospheric contamination monitors, and a variety of safety and alarm devices, shown in Figs. 116 to 121.

Technique in Surveying

When applying the inverse square law to gamma photons emanating from an unshielded point source, the dose rate can be estimated at any one point from the measurements taken at another. With beta sources, the emitted particles will have a spectrum of energies ranging from zero to some maximum energy, $E_{\max}$. The increase in radiation intensity found as one approaches a beta or a gamma radiation source is greater than would be predicted by the inverse square law. These phenomena were discussed in Chapter 5. The range of beta particles in the air is shown in Table 75.

Table 75

RANGE OF BETA PARTICLES IN AIR[1]

Energy (Mev)	Maximum Range of Beta Rays (Meters of Air)
.01	.0022
.02	.0072
.04	.024
.08	.080
.15	.21
.20	.36
.40	1.0
.8	2.8
1.5	6.1
3.0	13.0
5.0	19.0

[1] From Moteff (1954).

The dose rate at one foot distance from a point source gamma irradiator is shown in Table 76. For unshielded sources which are both gamma and beta emitters, it is possible to estimate the dose rate due to the respective radiation components. By closing the window on the Geiger counter tube, an indication of gamma flux alone is obtained. The open window reading will indicate the flux due to both beta and gamma. Subtracting the first from the second yields beta radiation intensity. While extremely low energy gamma radiation may be absorbed by the window material, for these purposes the assumption is that the window is sufficiently thick to absorb just the beta radiation.

Air Monitoring

During routine processing when air contamination is possible, it is important to prevent respiratory contamination of personnel. One method of evaluation is to have air drawn through an electrostatic precipitator; the precipitate is then removed and counted in a suitable counter.

Table 76

CALCULATED DOSAGE RATES AT ONE FOOT FROM POINT SOURCE GAMMA EMITTER[1]

E (Mev)	Rads/hr. per Curie
0.1	0.5
0.2	1.1
0.3	1.8
0.5	3.1
1.0	6.0
1.5	8.2
2.0	10.0
4.0	15.0

[1] From Moteff (1954).

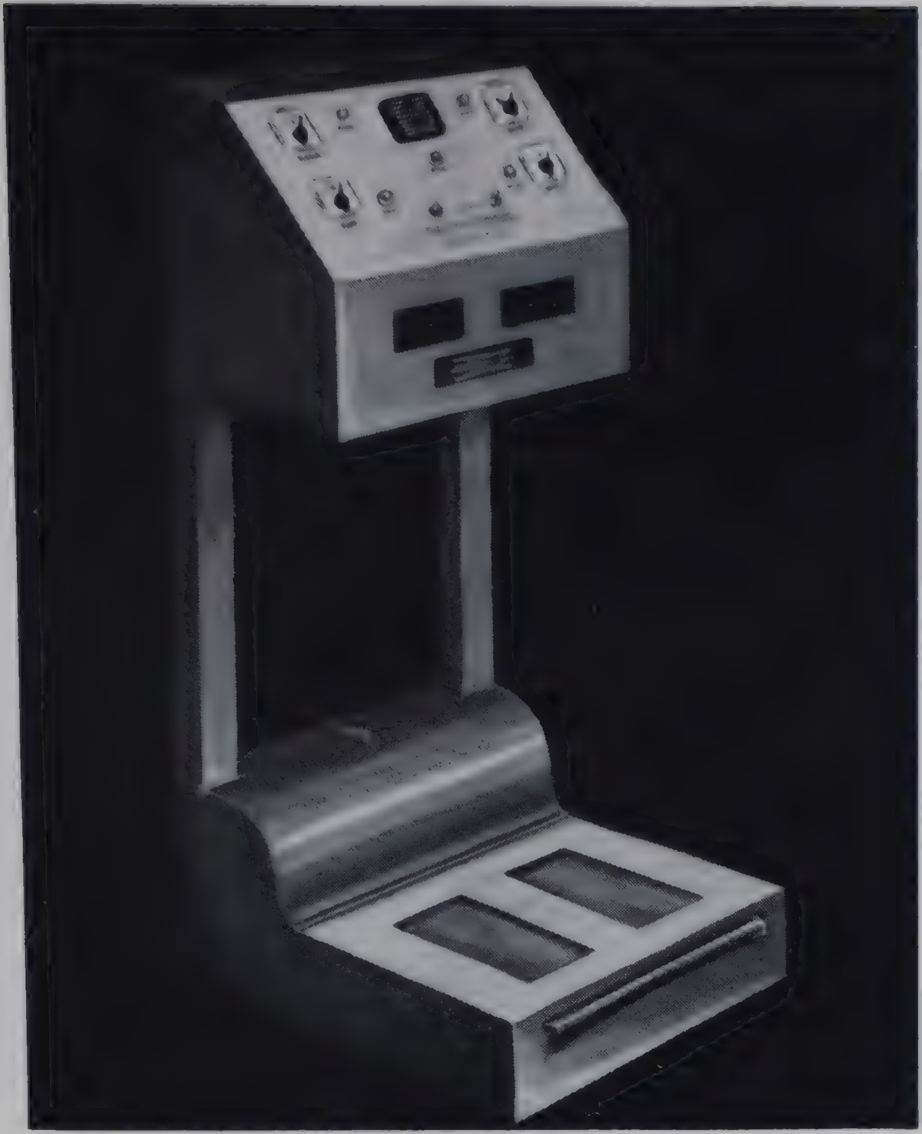


Fig. 118. Beta—Gamma Hand and Foot Monitor

Survey of Surfaces

In the survey of surfaces where beta and gamma contaminations may occur it is preferable to use instruments which have sensitivities greater than 0.1 mr per hour. Geiger-Mueller tubes are functional in detecting the presence of radioactive contamination. The response is fast and may be used for quantitative measurements following calibration. Some suggested limits for radioactive contamination are shown in Table 77. When contaminated material is uncovered in survey, it must be immediately isolated and controlled.

Personnel Monitoring

The routine monitoring of personnel may be accomplished by each individual wearing a suitable radiation detection device, preferably a device which can be worn on those parts of the body most likely to receive the maximum dose. Usually the radiation monitoring unit is worn

TABLE 77

ACCEPTED LIMITS FOR RADIOACTIVE CONTAMINATION[1]

Item	Maximum Permissible Contamination
Air	10^{-11} mc./cc.
Water	10^{-7} mc./cc.
Clothing	0.1 mr./hr. or equivalent
Work area surfaces	0.1 mr./hr. or equivalent
Body surfaces	0.05 mr./hr. or equivalent
Smear test on table tops from equipment (2 sq. in. filter paper smeared over an area of 12 sq. in.)	0.05 mr./hr. or equivalent

[1] From Morgan (1947).

on the chest or at the waist. There are several types of radiation monitoring devices suitable for personnel monitoring including ionization pocket meters and film packets. One or more of these meters can be worn by all personnel. It has been found desirable to have monitoring devices worn by personnel even when very low levels of radiation are to be encountered. The accidental or unknowing exposures must be controlled and identified for personnel protection.

Monitoring of Hands, Feet, and Clothing

Hands and feet should be monitored immediately when opportunity of contamination occurs. Special counters have been developed which permit the checking of hands and feet simultaneously. Such units should be located near the exits from radiation areas and also near wash rooms and dressing rooms. Frequent scanning of clothing for contamination is indicated.

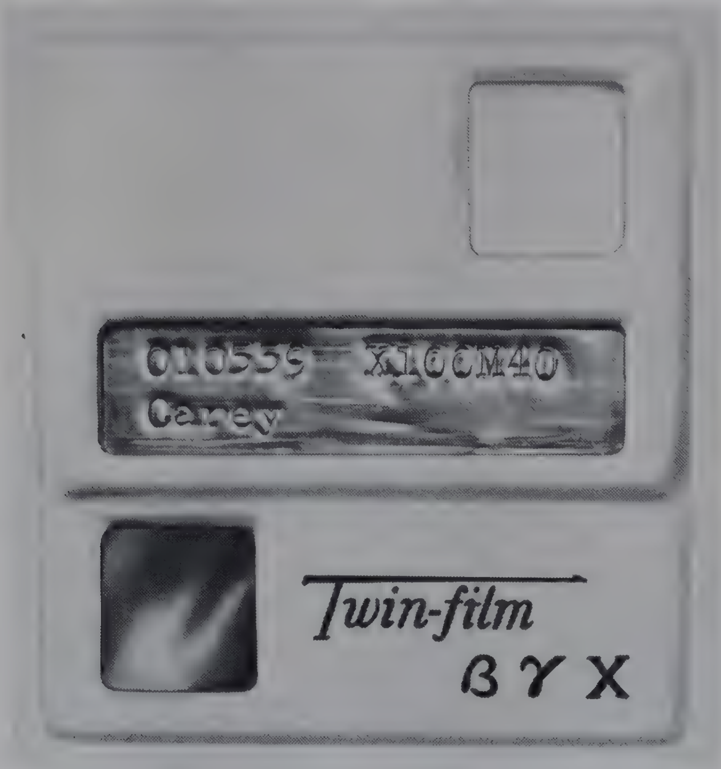
Maintenance of Adequate Records

Personnel monitoring data must be retained as a permanent record for all personnel. Survey records from operations and from the atmosphere in the installation should be a part of the permanent record.

All personnel operating within a nuclear radiation facility whose safety depends upon proper operation rather than strictly on shielding should wear one or more monitoring devices at all times when exposure is possible. The film badge is perhaps the most widely used and most successful of the personnel dosimeters. A number of commercial companies handle film badges as a service. It is a constant problem with personnel working with radiation to have them adhere closely to established safety regulations. Because radiation effects are generally delayed, some laxity develops due to overconfidence. If accidents are to be avoided, this cannot be permitted. Safety methods and recommendations listed in reference have been developed to the point where it is unnecessary for anyone to suffer ill effects from working with radiation. Like fire, radiation must receive its due respect. We must not permit the same degree of irresponsibility with radiation that has been tolerated with fire.

Radiation Shielding

The most obvious means of protecting personnel from radiation is to keep them away from it. This may be accomplished in a facility by increasing the distance between the operator and the source, or by employing suitable absorbers or shielding materials which will prevent passage of radiation from the source to the personnel working in the area. The nature of the shielding required to insure the safety of personnel is naturally related to the nature of the radiations being encountered. Rubber gloves, for example, are adequate protection against alpha emitters, but great care must be exercised to prevent atmospheric contamination, and inhalation. Adequate ventilation is therefore an important consideration.

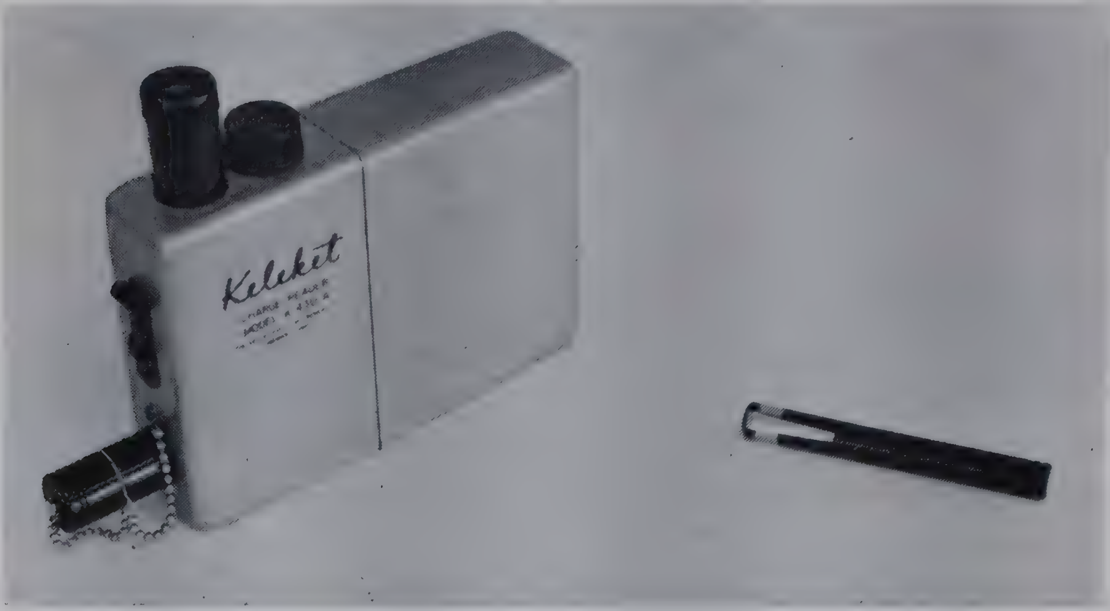


Courtesy of Tracerlab

FIG. 119. FILM BADGE—EACH UNIT CONTAINS TWO FILMS

One is sensitive for measurements from 0 to 2 rads. The other is less sensitive for dosages up to 30 rads.

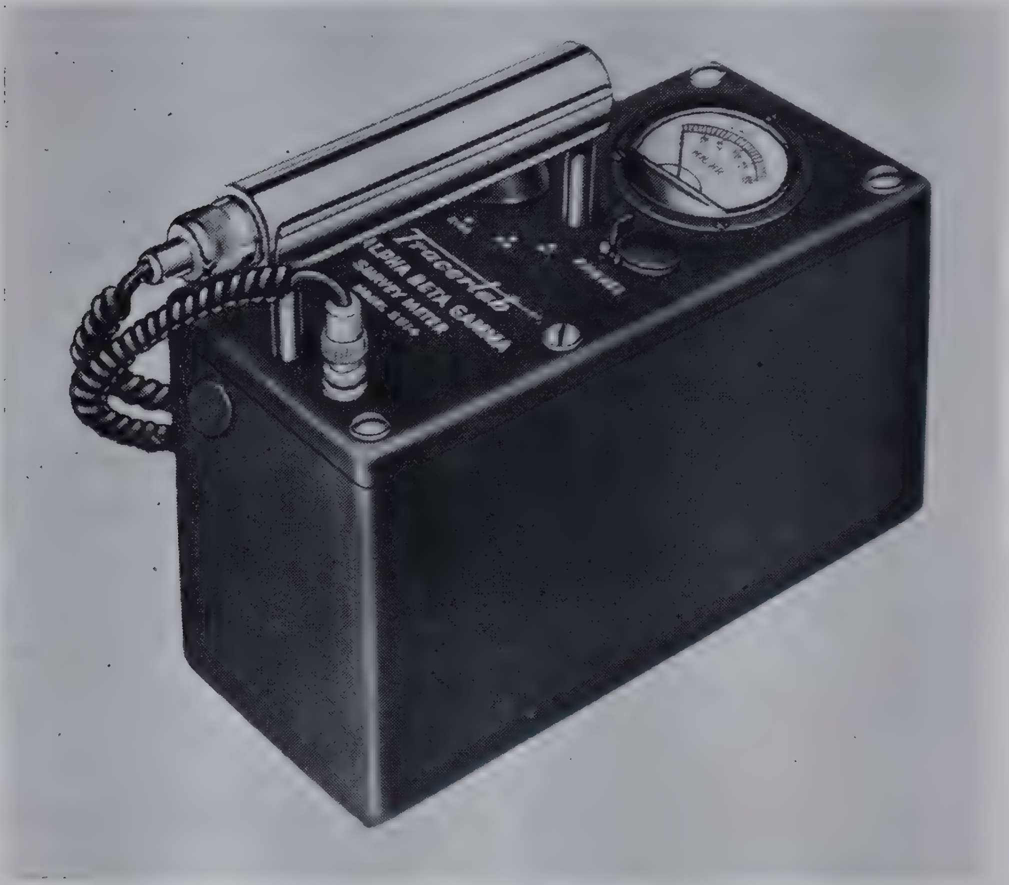
The thickness of the shielding required for a given high energy electron



Courtesy of Tracerlab

FIG. 120. POCKET DOSIMETER AND DOSIMETER CHARGER

Accurate measure of radiation absorbed during a given time period—the accumulated dose. Range from 200 millirads to 10 rads.


Tracerlab
ALPHA BETA GAMMA
SURVEY METER

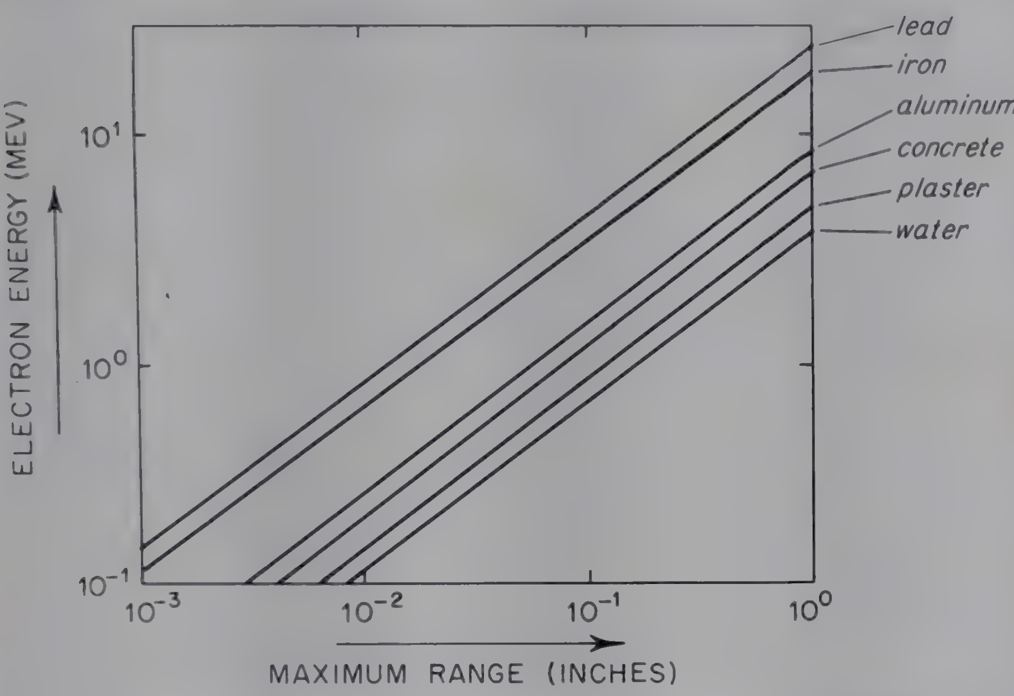
Courtesy of Tracerlab

FIG. 121. ALPHA, BETA, GAMMA SURVEY METER

A Geiger tube instrument with long battery life. Ranges 0.25 to 25 millirads per hour. Movable probe.

source decreases with increasing density of the shielding material. This was shown earlier to be related to the concentration of electrons per unit volume of the material. However, the use of heavy metal shielding materials is not recommended because of the problem of secondary radiation, which can be a greater problem than primary radiation. In practice low atomic weight materials are used for shielding beta radiation, graphite or concrete blocks, water, soil, glass, and plastics being suitable. The relative efficiency of several materials in stopping high energy electrons is shown in Fig. 122.

With gamma rays and neutrons, due to their great penetrating power in matter, a more difficult problem arises. The best absorbers of gamma radiation are the high density materials. Hence, lead, which has a high electron density, is one of the best shielding materials for gamma radiation. Due to its cost, however, it is useful to employ, where possible, great thicknesses of soil, concrete, or water, which are inexpensive and widely available. This can be done where size is not a limiting factor. Concrete is perhaps the most widely used neutron shielding material.

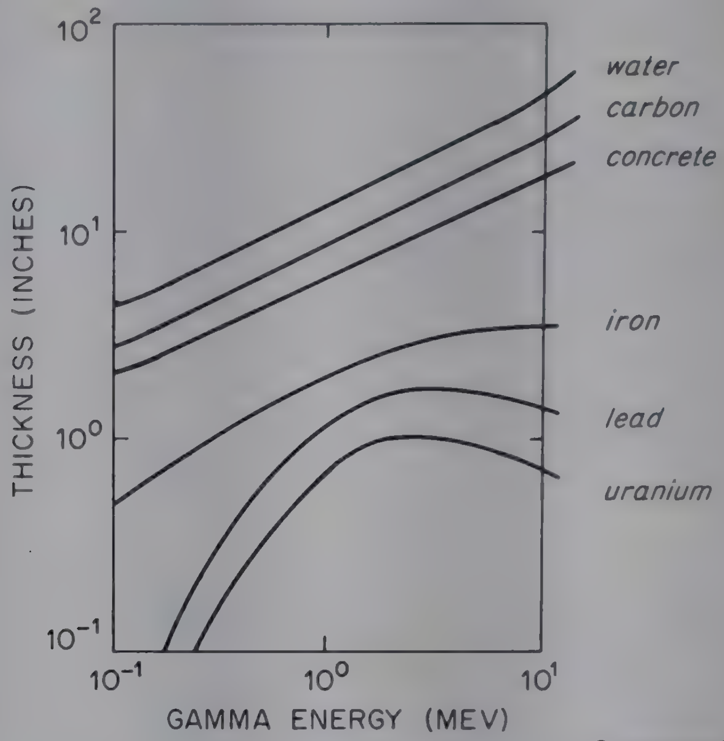


Courtesy of Moteff

FIG. 122. SHIELDING REQUIREMENTS FOR HIGH ENERGY ELECTRONS IN SEVERAL MATERIALS

Spent fuel elements from reactors are easily shielded in great depths of water to permit cooling from a radiation standpoint before being handled by remote control means. Movement of fuel elements from cooling canals to irradiation areas is accomplished by using remote control systems, placing the intensely radioactive elements in huge lead chests, and moving the chest. The stopping power of a number of materials for gamma radiation is shown in Fig. 123.

In final analysis the problem of satisfactory shielding of radiations is a direct application of the information gained concerning the interaction of radiation with matter (Chapter 5).



Courtesy of Moteff

FIG. 123. SHIELDING REQUIREMENTS FOR GAMMA RADIATION TO ATTENUATE BY A FACTOR OF TEN

Federal Regulations

Information concerning the federal regulations governing the procedures and requirements for operating radiation sources can be obtained by writing to The Director, Division of Civilian Application, The United States Atomic Energy Commission, Germantown, Maryland. Selected references covering the subject matter are also listed in reference.

Contamination of Air, Water, and Earth With Radioactive Elements

It is now clear that increasing the intensity of radiation above that normal to the earth has significant manifestations on all life. Therefore, it is important to consider the effects of contamination of whole masses of land which could result from nuclear explosions, accidents from nuclear reactors, and from fires in industrial plants which contain large amounts of radioactive materials.

In the ordinary operation of a nuclear reactor, the control of radioactive effluents is an important problem which has been satisfactorily solved.

While there was no past experience to act as a guide in this matter, progress in nuclear engineering was paced by progress in waste disposal. Waste disposal follows two patterns—concentration and isolation, or dilution (Glasstone 1958).

By far the greatest amount of radioactive materials produced without accidents or explosions comes from the operation of reactors and chemical separation plants. The radioactive materials produced can be solids, liquids, or gases. The gases result from the cooling of reactors with air and from gases liberated by the production and subsequent treatment of fission products. The neutrons inherent in a reactor activate the air, and radioactive nitrogen, oxygen, carbon, and argon are produced. The nitrogen and oxygen radioisotopes have very short half-lives, less than half a minute, hence are a minor problem that can be overcome simply by delaying their release. Carbon-14 is yielded in rather small amounts; so radioactive argon-41 is the critical component, and it has a half life of about two hours. In addition, some radioactive dust may be carried off accidentally in the air.

Consequently, as the air is removed from the reactor, it is passed through precipitators and filters and then discharged through tall stacks into the atmosphere. The air surrounding such installations is constantly monitored, and no release occurs when atmospheric conditions are not favorable. The great dilution of the gases in the atmosphere reduces the problem to insignificance.

When uranium fuel is reprocessed as the first stage in the extraction of plutonium, radioactive gases are evolved. Gaseous fission products are also liberated, notably radioactive iodine and xenon. These gases are carried away in large volumes of air, any suspended radioactive dusts are removed, then the cleaned air is released from tall stacks to be diluted in the atmosphere.

Liquid wastes from reactors are of two types, the water used to cool reactors, and the water used in chemical reprocessing plants. In the first case, the water exposed to high neutron concentration yields stable isotopes of hydrogen and oxygen, but radioactive elements are made from the salts dissolved in the water itself. Such cooling water can be held in lagoons to allow radioactive decay processes to operate, followed by the release of the stored water in small volumes into large volumes of water, dilution again operating in our favor. However, at times the cooling water can become very radioactive by picking up fission products from ruptured fuel elements. By monitoring the water, precipitating the harmful contaminants and filtering, partially purified water can be released into large rivers after a suitable period of holding.

The water from chemical reprocessing plants presents another type of problem, as this water must be treated. In general, this liquid waste is concentrated by any of several methods so that a reduced bulk of highly radioactive material is obtained, then the separated water is purified and discharged into rivers. The sludge may be held for two or more years in large lagoons until the activity of the waste is reduced sufficiently to permit its further treatment and release.

The solid waste materials are either packaged and buried in the ground or dumped at sea. Contaminated wood and other combustible matter is burned and the ashes are buried. The gases discharged in such flames must be controlled, as noted above.

But what happens when all the well detailed plans are rendered useless by accidents, explosions, and fires? It is necessary to anticipate the contamination of the atmosphere, of water supplies, and of soil.

Our atmosphere is being monitored all over the earth at this moment. The waters of the earth are being monitored, regularly. Our food supply is being monitored with less frequency.

Air can be filtered and purified without too complicated a system in enclosures, such as a building. Water supplies can be purified by ion exchange techniques similar to those used in water softening. Uncontaminated water reserves can be maintained for some period following an accident or contamination. But what about the earth's surface?

A main channel through which radioactive material can pass into living organisms is through depositions in soils, to be taken up by plants, then ingested by animals and men.

It is the cyclic nature of the life processes which has great significance to us because of the passage of radioactive elements from the air and water to soil to vegetation to animals to humans. It was noted earlier that man consumes ten times his body weight in food per year, and perhaps some 1,500 times his body weight at adulthood in the course of a 70 year life. The isotope strontium-90 is then a major concern. Next in importance is cesium-137, but since its rate of absorption from the soil is less than that of strontium (due to the substitution possibility of strontium for calcium), strontium remains the major hazard.

It is important to separate two situations, first, the deposit of radioactive elements on land in which crops are growing, and second, the contamination of bare land which can be manipulated before crops are sown. In the former case contamination may be distributed between leaves and stems and roots and in the soil's surface. In the latter situation strontium-90 can be distributed for some distance into the soil depths.

Absorption from the soil has received some study. Russell (1958) applied strontium-89 and 90 as sprays on the surface of land in which plants were growing or seeds were germinating, and alternately, applied strontium to bare lands in which crops were to be cultivated by various procedures.

The calcium content of soils is regarded as the major soil factor which can be altered which will affect the absorption of strontium. It is well established that in calcium deficient soils, the relation of strontium to calcium entering the plants is enhanced possibly by a factor of three or more.

It has been found that plants differ somewhat in their ability to absorb strontium relative to calcium from soil, particularly the root crops, which in general absorb greater amounts (Russell 1958). However, there are several major sources of calcium in the dietary, and roots generally supply but a small fraction of the calcium requirement in humans.

In cultivated soils in England it has been found that about one per cent of the strontium which is applied to a field is absorbed by a crop in any one season (Russell 1958). The average ratio for strontium-90 to calcium absorbed in good soils appears to be about one micro-microcurie of strontium per gram of calcium, if the deposition is one millicurie per quarter mile square.

The hazard which exists when widespread distribution of radioactive elements occurs obviously depends on many factors, especially the nature, the abundance, and the rate at which the radioactive material can move through the pyramid of life.

The radioisotopes of iodine and strontium are accumulated in the thyroid and in the bones respectively. These are the two radioactive elements most likely to give rise to hazards through contamination of the earth and its atmosphere from nuclear reactions. For practical purposes it may be sufficient to consider just these two elements.

Since the passage of these elements through soils and plants is relatively slow, a short term effect is to be recognized. When dealing with short-lived radioactive materials such as iodine-131 with a half-life of eight days, an immediate hazard from all accidents is due to such elements being deposited through water and air supplies. But the longer-lived radioactive elements, for example strontium-89 (half-life 53 days) and strontium-90 (half-life 27 years), especially the latter, involve a long chain of events and have significant long term ramifications. When any deposition occurs, it is necessary to consider also the hazard from the inhalation and external radiation. Radiation effects were discussed earlier.

What action is necessary after contamination to recover the earth's surface for agricultural purposes? If assumptions are made regarding the amount of plant growth in the soil or the dietary habits of the animals feeding from it, it is possible to calculate the probable contamination of milk, for example, which will result from a fallout per unit area of ground surface or per unit of vegetation on the earth. But such calculations are most approximate, since local conditions can cause the relationship to vary greatly. For example, it is thought that in a heavy rain or with heavy irrigation, the strontium which is deposited will be removed from the vegetation more rapidly than the iodine, which may be simultaneously deposited. Moreover, particularly in the early stages, the relation between the amount deposited and the level found in milk may vary greatly depending again on the size of the dust which falls. This being true, the levels of contamination in milk and other articles of food which are consumed by humans can be determined satisfactorily only by direct assay of the food after hazardous deposition.

From what has been learned, the replanting of fields brings about a considerable reduction in hazard, possibly by a factor approaching 10. With the shallow rooted crops and some grasses, deep cultivation may reduce the resultant strontium-90 content in plants by a factor of the order of three. When shallow rooted plants are grown for a crop, cultivation a foot and more in depth is particularly beneficial. A significant amount of strontium will be turned below that zone from which the plant absorbs directly and actively. This effect will be improved with the coming of uncontaminated rains, or by irrigation. The deep-rooted species of plants, in which all our cereal crops are classified, pose a different problem. With deep rooted species, the effect of cultivation is the opposite.

If the nature of the soil in which plants are growing is such that the plants can establish a very deep root system, for example, corn penetrating to eight or more feet, there appears to be an advantage in drawing nutrients from immediately uncontaminated soil. Lucerne grass in particular is useful because it absorbs calcium selectively, leaving behind the strontium.

Even if the methods available for reducing the ratio of strontium-90 to calcium in plants are not practicable or inadequate, a further opportunity for protection arises. That is, the pattern of agriculture and plant growth can be altered so that the products which contribute little to the dietary calcium of mankind are produced. The restriction of the diets of dairy herds is an obvious control.

Furthermore, shallow-rooted crops can be planted which are harvested and destroyed. The filtering effect which such new growths have is useful in depleting the soil surface of its radioactive contamination. It must be emphasized that the information to permit man to live effectively in once contaminated land is incomplete at present.

The exact conditions at the time of contamination will cause wide variation in the hazards which result from a deposit by radioactive rainfall.

dust, or accident. However, sufficient is known to identify milk from pastured cows as a food likely to show the widespread contamination at an early stage; and the probable time of transfer of some of the fission products to milk is but a few days.

With appropriate organization and appropriate monitoring techniques, it is thus possible to give a society of mankind a degree of protection in the event of radioactive contamination of the earth and its atmosphere that compares favorably with protection from less spectacular but more familiar types of industrial accidents.

In summary, there are two fundamental hazards associated with radioactive rainfall and dust. It is essential that ingestion and inhalation of radioactive material be prevented. Protection against the entry of radioactive materials into the body can be obtained by maintaining vigilance, with effective contamination control methods and by immaculate techniques with rigid systems of regulation. While the present levels of radiation and radioactive contamination of the earth have increased slightly by experimentation with nuclear energy, the levels are thought to be below those which can inflict widespread damage to mankind. It has been estimated that the level is less than one per cent of the critical level. However, even slight increases over the natural radiation background can have measurable effects on living organisms, although the manifestation may be delayed for many generations.

Perhaps we shall learn something of the background effects themselves in time. This suggests that, while there is no reason for alarm currently, there is reason for careful thought and planning for the future.

We must participate in nothing less than an adequate effort to prevent the accidental irradiation of mankind in general. But this need not retard the exploitation of atomic energy for the benefit of mankind.

BIBLIOGRAPHY

British Institute of Radiobiology. 1955. Recommendations of the Intern. Commission on Radiobiological Protection. Supplement No. 6. London, England.

Butts, J. S. 1958. Position and trends in O.E.E.C. member countries on the applications of atomic science. E.P.A.-O.E.E.C. Meeting, Application of Atomic Energy in Agriculture and Food, Paris, France.

Claus, W. D. 1958. Radiation Biology and Medicine. Addison-Wesley Publ. Co., Inc., Reading, Mass.

Cooper, R. D., and Siu, R. G. H. 1957. Radiological safety, Chapter 31. Radiation Preservation of Food. U. S. Govt. Printing Office, Washington, D. C.

Federal Register. 1956. Title 10. Atomic Energy. Part 30—Licensing of by product material. Federal Register of the United States, *31*, 213–218, Jan. 11, 1956. Washington, D. C.

GLASSTONE, S. 1958. Sourcebook on Atomic Energy. Second Ed. D. Van Nostrand Co., Inc., New York, N. Y.

HURSH, J. B., ZIZZO, S., and DAHL, A. H. 1951. Survey meters for water monitoring. Nucleonics 9, No. 5, C16–C20.

KUPER, J. B. H., and COWAN, F. P. 1958. Dosimetry and instrumentation for hazard evaluation. Chapter 18. Radiation Biology and Medicine. Addison-Wesley Publ. Co., Inc., Reading, Mass.

LETAVET, A. A. 1955. Health protection of workers exposed to ionizing radiations (in Russia). Proc. Intern. Conf. Peaceful Uses of Atomic Energy *13*, 159–161.

LOUGH, S. A., and SOLON, L. R. 1958. The natural radiation environment. Chapter 17. Radiation Biology and Medicine. Addison-Wesley Publ. Co., Inc., Reading, Mass.

MORGAN, G. W. 1948. Some practical considerations in radiation shielding. U. S. Atomic Energy Commission, Isotopes Div., Circ. B-4, Washington, D. C.

MORGAN, G. W. 1949. Surveying and monitoring of radiation from radioisotopes. Nucleonics *4*, No. 3, 24–37.

MORGAN, K. Z. 1947. Health physics and its control of radiation exposures at Clinton National Laboratory. Chem. and Eng. News *25*, 3794–3798.

MOTEFF, J. 1954. Miscellaneous data for shielding calculations. Rept. APEX 176. General Electric Company, Milwaukee, Wisc.

NATIONAL ACADEMY OF SCIENCE—NATIONAL RESEARCH COUNCIL. 1956. The biological effects of atomic radiations. Washington, D. C.

NATIONAL ACADEMY OF SCIENCE. 1956. Genetic effects of atomic radiation. Science *123*, 1157–1158.

OLSON, R. A. 1958. Radioisotopes in soil and plant research. E.P.A.-O.E.E.C. Meeting—Applications of Atomic Science in Agriculture and Food. Paris, France.

POLLARD, E., and DAVIDSON, W. 1951. Applied Nuclear Physics. John Wiley and Sons, Inc., New York, N. Y.

PURDUE UNIVERSITY. 1957. Radiological Control Handbook. Lafayette, Indiana.

RUCHHOFT, C. C., GORMAN, A. E., and CHRISTENSON, C. W. 1952. Wastes containing radioactive isotopes. Ind. Eng. Chem. *44*, 545–546.

RUSSELL, R. S. 1958. Agricultural aspects of contamination arising from the use of atomic energy. E.P.A.-O.E.E.C. Project 396. Paris, France.

SCHUFFELEN, A. 1958. Radioisotopes in soil research. E.P.A.-O.E.E.C. Meeting—Application of Atomic Science in Agriculture and Food. Paris, France.

STONE, R. S. 1952. The concept of a maximum permissible exposure. Radiology 58, 639–642.

UNITED KINGDOM MINISTRY OF LABOUR AND NATIONAL SERVICE. 1953. Precautions in the use of ionizing radiations in industry. Her Majesty's Stationery Office, London, England.

U. S. ATOMIC ENERGY COMMISSION. 1947. General rules and procedures concerning radioactive hazards. Isotopes Div., Circ. B-1. Washington, D. C.

U. S. ATOMIC ENERGY COMMISSION. 1950. The effects of atomic weapons. Combat Forces Press, Washington, D. C.

U. S. DEPARTMENT OF COMMERCE. 1949. Safe handling of radioactive isotopes. U. S. National Bureau of Standards, Handbook No. 42. Washington, D. C.

U. S. Department of Commerce. 1952. Radiobiological monitoring methods and instruments. U. S. National Bureau of Standards Handbook No. 51. Washington, D. C.

U. S. Department of Commerce. 1954. Protection against betatron synchrotron radiations up to 100 million electron volts. U. S. National Bureau of Standards Handbook No. 55. Washington, D. C.

U. S. Department of Commerce. 1954. Permissible dose from external sources of ionizing radiation. U. S. National Bureau of Standards Handbook No. 59. Washington, D. C.

U. S. Department of Commerce. 1955. Regulation of radiation exposure by legislative means. U. S. National Bureau of Standards Handbook No. 61. Washington, D. C.

U. S. Department of Commerce. 1953. Maximum permissible amounts of radioisotopes in the human body; Maximum permissible concentrations in air and water. U. S. National Bureau of Standards Handbook No. 52. Washington, D. C.

Index

A

Acanthoscelides obtectus, irradiation, 274
Accelerators, 75–81
  processing cost analysis, 365–72
Acetic acid, *see Acids*
Acetylene, irradiation, 116–7
Acids, irradiation, 124–5
Activated complex, 37, 40
Adelina deronis, irradiation, 271
Aedes aegyptia, irradiation, 272
Alcohols, irradiation, 119–21
  free radical mechanisms, 119–21
  methane yields, 121
  oxygen effects, 121
Aldehydes, irradiation, 123–4
Alpha particle, 10
  absorption, 14
  track, 55
Alveolar hypothesis, 174
Amino acids, irradiation, 128–31
  ammonia yield in, 129
  radiation synthesis of, 353
Antibiotics, radiation sterilization of, 346–7
  use in food sterilization, 329
Aqueous solutions, irradiation, 108–10
  oxidation-reduction reactions in, 108–10
Ascaris Lumbricoides, 268–70
  irradiation, 270
Ascorbic acid oxidase, heat destruction, 329
Asparagus, irradiation, 253
Aspergillus oryzae, irradiation, 222
Aspergillus tenuis, irradiation, 222
Atomic Energy Commission, 324, 333
Atomic number, 8, 13, 15
Atomic radiation, *see Alpha particles, Protons, Fission fragments*
Atomic radius of elements, 26
Auxin, changes in irradiation, 234–6
  in plant growth, 233

B

Bacillus coagulans, irradiation, 212, 215–17
  effect of biotin on irradiated, 215
  post irradiation recovery, 216
  post irradiation temperature sensitivity, 217
  radiation resistance of spores and vegetative cells, 212
Bacillus subtilis spores, irradiation, 213
  concentration effects, 213
Bacon, process for radiation pasteurization, 335–6
Bandages, radiation sterilization of, 346–8
Barley seed, irradiation, 254
Bean weevil, *see Acanthoscelides obtectus*
Beef, control of tenderness in, 308
  D value of *Cl. botulinum* in boiled, 214, 219
  effect of radiation on enzymes, 165
    flavor, 304
    glycogen content, 147
    niacin retention, 135
    PDR index, 303
    pigments, 138–9
    riboflavin retention, 135
    thiamin retention, 135
  enzyme inhibition by heat, 328–9
  induced radioactivity in, 53–4
  protein, amino acid composition, 286
  taenia saginata in, 268–9
Beets, irradiation, 254
Benzene, irradiation, 116, 130
  formation in irradiated acetylene, 116
Biotin, effect on irradiated spores, 215
Bologna, radiation sterilized, 308–9
Bond energy, 34–36
  ions, 35
  molecules, 35
Bone marrow, irradiation, 171
Botulism, 293–4
Bremsstrahlung, 46
  energy loss by atomic radiation, 49
    by electrons, 47–8
Broccoli, irradiated, 135
Butane, irradiated, 113–4

C

Cadelle, 275
Capacitron, 80
Carbon-14, 5, 387
  dating, 18
  half life, 17
  labeled methanol, self-irradiation, 120
  production by cosmic radiation, 5, 12, 17–18
  use to study photosynthesis, 181
Carbonates, aqueous solutions, irradiation, 125
Carotenoids, radiation stability, 132
Casein, irradiation, 158–9
Catalysts, irradiation, 354
Cathode rays, *see Electrons*
Cells, death of, 207
  dynamic state of, 178–9

Cells (*continued*)
  effect of temperature on shape, 179
  growth of, 189–91
  irradiation of, 205–6
    effect of age, 215
      atmosphere, 213
      concentration, 212–13
      oxygen, 208
      pH, 214
      radiation quality, 215–16
      species, 212
      substrate, 213–14
      temperature, 213–14
      vegetative *vs* spore state, 209–212
      water, 208–9
  metabolism, 179–91
  mitosis, 191–95
  nutrient requirements, 187–9
  respiration, 182–4
  structure, 172–8
Cell membrane, irradiation, 210
  structure, 185–6
  transport across, 184–7
Cellophane, irradiated, 316–17
Cellulose, irradiation, 146, 312, 315–16, 350
  grease proofness, 316
  irradiated plant, 253
  tearing resistance, 316
  tensile strength, 315-16
Cesium-137, 74
  gamma ray energy, 74
  half life, 74
  in medical therapy, 351
  in soil, 388
  processing cost analysis, 365–72
Chain reaction, 62–4
Chemical reactions, radiation initiation of, 38
Cherries, irradiation, 252–3
Chicken, control of rigor mortis, 308
  enzyme inactivation in, 336
  irradiated, 309–10
  irradiated, color of, 139, 336–7
  irradiated, storage of, 309
  process for radiation sterilization of, 336–7
  tenderness of flesh, 304, 308
Chromosomes, aberrations, 194, 198–200, 237–9
  irradiation, 203
  radiation effects on plant, 237–9
  structure, 197–8
Cigarette beetle, *see Lasioderma serricorne*
Clonorchis sinensis, 268–9
Clostridium botulinum, *D* value, 326
  D value in boiled ground beef, 214
    in cream of chicken soup, 214
    in pea puree, 214
    in whole kernel corn, 214
  effect of water on *D* value, 209, 214
  in food, 293–5
  radiation destruction of, in food, 295–6
  radiation resistance of spores, 212, 219
Cobalt-60, 75
  gamma ray energy, 75
  half life, 60
  in medical therapy, 351
  irradiation of plants with, 230–31, 243–55
  processing cost analysis, 365–72
  radiation processing capacity, 361–4
Compton effect, 49–51
Corn, *D* value for *Cl. botulinum* in, 214
  protein, amino acid composition of, 286
Cosmic radiation, 4–5, 375
Cream of chicken soup, *D* value for *Cl. botulinum* in, 214
Cucumber ascorbic acid oxidase, irradiated, 165
Curie, definition of, 18
Cyclohexane, irradiation, 116
Cyclohexene, irradiation, 116
Cyclotron, 5, 14, 75

D

D value, definition, 213, 226
  of aciduric bacteria, 296
  of *Cl. botulinum*, 212, 214–5, 219, 295–6, 326–7
  of enzymes, 327–8
  of *E. coli*, 213
  of molds, 296
  of viruses, 225
  of yeasts, 296
Death, problem of, 207
Deoxyribonucleic acid, 167–8
  and cell division, 191
  and genetics, 177–8
  irradiation of, 167–8, 202–4, 236–7
  structure of, 195–7
Deuteron, binding energy, 10
  mass, of, 10
Diffusion in solids and liquids, 32–33
Diphyllobothrium latum, 268–9
Disaccharides, irradiation, 127–8
Dose, radiation, *see Radiation dose*
Dosimeter, 91–95
  and health physics, 97–98
  ceric sulfate, 91, 93, 109, 335
  cobalt glass, 91–94, 335
  ferrous sulfate, 91–2, 108–9, 333–4
  film badge, 383
  go-no-go, 94–5
  pocket, 384
  primary standard, 91
Drosophila melanogaster, irradiation, 273–4
Dugway Proving Ground Radiation Facility, 361
Dynamitron, 80–81

E

Egg albumin, irradiation, 158–9
  radiation protective action of, 164–5
Eimeria tenella, irradiation, 271
Electricity, production from radioisotopes, 354
Electron, affinity of elements, 26
  high energy, dose-depth relation, 78
    machine generated, process economics, 365–72
    maximum range of, 97, 380
    penetration of, 51, 78, 83–5, 330
    radioisotope source, process economics, 365–72
    scattering of, 48
    shielding requirements, 385
    tracks, 54–55
  mass, 8
  pair bond, 28–9
  secondary, ejection by high energy electrons, 46–7
    energy distribution of, 47
  spin, 23, 29
  trapping substances, 115
  wave functions, 22
Electron-Positron pair production, 49–50
Electron volt, 9
Endamoeba histolytica, 271
Enzymes, *D* value of, 327–8
  effect of radiation on, 161–7
  effect of radiation on reactions of, 165–6
  effect of temperature on radiation sensitivity, 165
  heat inactivation of, 166, 329
  nature of, 159–61, 179–80
  plant, radiation effects on, 235–7
  production of sterile, 354
  radiation resistance of dry, 166
  target molecular weight of, 163
Epinephrine, 329
Escherischia coli, effect of biotin on irradiated, 217
  irradiation, 212
  nutrient requirements, 187
  post irradiation recovery of, 217
  post irradiation treatment of, 217–19
Ethers, irradiation, 121–3
  gaseous hydrocarbons yields in, 122
  olefin yields in, 122
Euplotes, irradiation, 272
Excited molecules and ions, decomposition of, 39–40
  spatial distribution of, 54–6

F

Fasciola hepatica, 268–9
Fasciolopsis buski, 268–9
Fats, irradiation, 125–7
  undesirable flavors in, 127
Fish, process for radiation sterilized, 336–7
  radiation treatment of, 329
  useful storage life of, 290
Fission, 15, 61–4
  discovery of, 61
  energy, distribution of, 62
  energy equivalent of, 61
  fragments, energy of, 72
  products, 15, 72–4
    kinetic energy of, 62
    radiation from, 86
  yield curve, 14
Flour, dose distribution by gamma rays in, 84
  dose distribution by high energy electrons in, 84
  irradiation, 276
  process for insect deinfestation, 333–4, 337
Flour beetle, *see Tribolium*
Food, distribution of, 287–8
  essential, table of, 288, 303
  irradiated, acceptance of, 306–10
    test program, 301–3
    wholesomeness of, 300–6
  poisoning, 291
  radiation processing of, 296–7
    combination treatment, 328–9
    general process requirements, 325
    increased storage life from, 331
    potential world market, 369–70
  recommended daily diet of, 284
  spoilage, 288–90
  spoilage organisms, 292–5
    radiation destruction of, 295–6
  useful storage life of, 290
Formic acid, *see Acids*
Free radicals, 34
  in irradiated aqueous solutions, 108–10
  in irradiated butane, 113–14
  in irradiated water, 103–6
  in oxidation of hydrocarbons, 117–18
  reactions of, 40–42
Fruit fly, *see Drosophila melanogaster*
Fruits, control of texture, 308–9
  irradiation of, 249–53, 337
    effect on maturation, 250–51
    effect on respiration, 249–50
    effect on storage stability, 251–3
  process for radiation pasteurization of, 334–5
  surface sterilization of, 251
  useful storage life of, 290
Fuel elements, spent, 73
  as gamma source, 95–6, 359–60
  radiation facility, 364

G

G value, 105
Gamma rays, 12–14
  dose-depth relation, 78

Gamma rays (*continued*)
  dose from source of, 380
  emission of, 14
  interaction with matter, 49–51
  shielding requirements, 386
  tracks of, 55
Geiger counter, 5
Glycine, *see Amino acids*
Glycogen, irradiation, 147–8
Grain beetle, *see Oryzaephilus surinamensis*
Grain borer, *see Phizopertha dominica*
Granary weevil, *see Sitophilus granarius*
Granular hypothesis, 174

H

Hadrobracon, irradiation, 273
Half lives of radioisotopes, 17
Ham, radiation sterilized, 307, 9
Health physics procedures, 333, 334
Helminthic diseases of man, 267–9
  control of, by radiation, 271
Helminths, radiation destruction of, 269–271
Heterodera, irradiation, 270
Hexane, irradiation, 112–13
Hydrocarbons, formation of peroxides in, 117–18
  irradiation, 112–19
    protective effect of benzene in, 116
  olefin dimerization, 115
  radiation stability of aromatic, 116
  radiolytic oxidation, 117–18
  scavenger reactions in irradiated, 113–14
Hydrogen bond, 146, 151, 211

I

Induced radioactivity, 52–4
  by electrons, 330
  by gamma rays, 82–3
  by neutrons, 52–4, 71–2
  in bacon, 335
  in beef, 53–4
  threshold energy for, 53
Insects, dose requirement for destruction, 273–6
  irradiation, 272–7
  penetration of packages by, 274–7
Insulin, irradiation, 155
  structure of, 150
Iodine-131, 17
Ionization, inner shell, 27
  outer shell, 27
  potential, of elements, 25
    of molecules and radicals, 43
Ion-molecule reactions, 44
  in liquid hydrocarbons, 115
Ions, reactions of, 42–44
  spatial distribution of, 54–6

K

Ketones, irradiation, 123–4
Kraft paper, irradiation, 315–16
Krebs citric acid cycle, 182–4

L

Lactic acid, *see Acids*
Lasioderma sericorne, irradiation, 274
Leather, radiation sterilization of, 349–50
Lethal radiation dose for living entities, 259
Linear accelerator, 5, 14, 77–9
Liquid cage, 32

M

Mass number, 8, 15
Meal worm, *see Tenebris molitor*
Median lethal dose, definition, 260
  for mice, 261
Milk, effect of heat on, 158–9
  effect of radiation on, 158–9
  flavor changes in irradiated, 159
  PDR index of, 303
  radiation inactivation of alkaline phosphatase in, 165
  rennet coagulation time in, 158–9
Moderator, neutron, 63–7
Molds, irradiation, 221–2
Molecules, geometry of, 29–32
  motions of, 32–4
Monosaccharides, irradiation, 127–8
  oxidation of, 127–8
  photosynthesis of, 180–2
  radiation synthesis of, 353
Mushroom tyrosinase, 165
Mutations, 194–200
  in animals, 266–7
  radiation induced, 202–4, 228
  useful plant, 254–5
Mylar, irradiation, 313–14, 319

N

National Academy of Science, 301, 329
National Bureau of Standards, 333, 335, 377
Nautilus, USS, 70
Neurospora, irradiation, 204
Neutrino, 13
Neutron, absorption, 14
  capture, 15, 61–4
  emission, 14
  fast, 63–4
  food sterilization by, 71–2
  mass of, 8, 10, 14
  moderator, 63–7
  reflectors, 64
Nuclear shell model, 10
Nuclear binding energy, 10
  reactions, 15
  structure, 8–9

- Nucleoproteins, 223
  - irradiation, 167–8, 202–4, 236–7
- Nylon, irradiation, 312, 314, 317

O

- Oats, irradiation, 276
- Oils, irradiation, 125–7
  - of natural, 350
  - undesirable flavors in, 127
- Ointments, radiation sterilization of, 346–7
- *Oryzaephilus surinamensis*, irradiation, 274

P

- Packaging materials, compatibility with radiation processing, 332
  - irradiation of, 311–22
  - radiation absorber characteristics of, 321
- *Pandorina morum*, irradiation, 272
- *Paragonimus westermani*, 268–9
- *Paramecium*, irradiation, 272
- Parasites, irradiation of animal, 267–72
- Pauli exclusion principle, 22
- Pea puree, *D* value for *Cl. botulinum* in, 214
- Peas, irradiated, chlorophyll in, 140
  - starch in, 253
  - sugar in, 253
- Pectin, irradiation, 147–8
- Pepsin solution, irradiation, 164
- Periodic table, 24–7
- Photoelectric effect, 49–53
  - threshold energy for, 53
- Photosynthesis, 180–83
- Pigments, animal, irradiation, 138–9
  - plant, irradiation, 140–41
- Plants, growth hormones, 233–4
  - growth of, 232–4
  - growth zones, 232–3
  - irradiated, 231–55
    - auxin levels in, 234–6
    - budding in, 240–2
    - growth of, 243–9
    - morphological changes in, 239–42
    - radiation damage sequence in, 231–2
    - tumors in, 240–2
  - strontium-90 in, 390
- *Plasmodium gallinacium*, irradiation, 271
- *Plasmodium lophorae*, irradiation, 271–2
- Pliofilm, irradiation, 315
- Plutonium-239, 66–71
- *Polmyxa*, irradiation, 272
- Polyethylene, irradiated, 312–14, 317–19
  - odor of, 313, 319
- Polymerization, 40, 42, 350
  - graft, 350
- Polymers, synthetic, irradiated, 311–22
  - gaseous products in, 315, 318
  - odors of, 319
  - package performance of, 314
  - stability of, 312
- Polystyrene, irradiated, 312–14, 319
- Pork, irradiated, color of, 139
  - niacin retention in, 135
  - PDR index, 303
  - riboflavin retention in, 135
  - thiamin retention in, 135
  - *Taenia solium* in, 268–9
  - *Trichinella spiralis* in, 268–9
- Positron, 13
- Potato, sprout inhibition by radiation, 243–9
  - dose requirements for, 244–9, 331–3, 337
  - process for, 331–3
  - storage characteristics of, 309, 333
- Protein, amino acid composition of, 286
  - denaturation, 32, 152–9
  - irradiation, 154–9
  - protection of irradiated viruses, 225
  - structure, 31, 149–54
- Proton, mass of, 8, 10
- Protozoa, irradiation, 271–2

Q

- Quantum mechanics, 20–3
  - and molecular structure, 28–31
- Quantum numbers, 21–23
- Quartermaster Food and Container Institute, 324

R

- Rad, *see Radiation dose*
- Radiation, definition of, 3
- Radiation dose, distribution across container, 95–97
  - Herschman equation, 96
  - gram roentgen, 90
  - lethal, 259
  - maximum permissible, 376–8
  - measurement, 89–98
  - median lethal, 260
  - rad, 91
  - roentgen equivalent mammal (rem), 90
  - roentgen equivalent physical (rep), 90
- Radiation monitoring systems, 378–83
- Radiation processing, capacity and source strength, 361–4
  - economics, 361, 365–72
  - plant, 362–5
- Radiation sickness, 375–6
- Radiation sources, general requirements for, 85–87
  - for food processing, 330–31
- Radiation sterilization, 225–27
  - of antibiotics, 346–7

Radiation sterilization (*continued*)
  of bandages, 346–8
  of food, process specifications, 338
  of fruits, 251–3
  of leather, 349–50
  of ointments, 346–7
  of plastic containers, 322
  of sewage, 351
  of sutures, 346–7
  process for, bacon, 335–6
    chicken, 336–7
    fish, 336–7
    flour, 333–4
    fruits, 334–5
    potatoes, 331–3
    vegetables, 336–7
Radiationless transition, 27
Radioactive contamination, limits, 382
  of air, 386–7
  of soil, 388–91
  of water, 387–8
Radioactive tracers, 120, 181, 352
Radioactive waste, treatment of, 64, 386–8
  utilization, 354
Radioactivity, 11–15
  half life, 16–18
Radioisotopes, preparation of, 13
  properties of, 17
Radish seeds, irradiation, 253
Radium, 5
Reactor, breeder, 71
  Calder Hall, 69
  control, 64
  dual purpose, 68–9, 370–71
  fast, 66–7
  homogeneous, 68–70
  Los Alamos fast, 69
  Materials Testing, 65–9, 360
  power, 67
  pressurized water, 69
  swimming pool, 68–9
  thermal, 65–7, 70
Rem, *see Radiation dose*
Rep, *see Radiation dose*
Resonant transformer, 79–80
Reticular hypothesis, 174
Rhizopertha dominica, irradiation, 274
Rice weevil, *see Sitophilus oryza*

S

Saran, irradiated, 312, 316–17
Savannah River Food Irradiation Facility, 359
Scavenger reactions, in acetone, 124
  in butane, 113–14
  in water, 105–6
  iodine in, 113
Scotchpak, irradiated, 314, 337
Screw worm fly, eradication, 277
Seeds, irradiation, 253–4
Sewage, radiation treatment, 351
Shielding, radiation, 64, 69, 81, 383–6
Shrimp, radiation sterilized, 305, 309
Sitophilus granarius, irradiation, 274–5
Sitophilus oryza, irradiation, 274
Sodium, liquid, as reactor coolant, 71
  loop radiation source, 71–2, 75, 371
Soil, radiation sterilization of, 350
  radioactive contamination of, 388–9
  treatment of radioactive, 389–91
Solar radiation, 3–5
Spices, irradiation, 276
Spinach, ascorbic acid retention in irradiated, 135
Spores, bacterial, radiation resistance of, 219–21
  influence of moisture on, 209
  resistance compared to vegetative form, 209–12
Staphylococcus aureus, irradiation, 214
Starch, irradiation, 146–7
  radiation effect on content in plants, 253
Strawberries, economic loss due to rot, 252
  irradiated, pigmentation of, 141
    storage stability of, 252
  storage of, 297
  surface sterilization of, 252
Strontium-89, 388–9
Strontium-90, and bone tumors, 266
  beta ray energy, 74
  half life of, 74
  in soil, 388–91
Surgeon General, U. S. Army, 300–301, 324
Sutures, radiation sterilization of, 346–7
Synchrocyclotron, 14

T

Taenia saginata, 268–9
Taenia solium, 268–9
Tareet theory, 162–3, 204–5, 224
Tenebria molitor, irradiation, 274
Termites, 275
Tobacco leaf, irradiation, 243
Toluene, irradiation, 116
Tomato, irradiated, pathogen susceptibility of, 251
  pigmentation of, 140, 249–50
  respiration rate of, 249–50
Trandescantia paludoso, irradiated, budding of, 240–42
Tribolium, irradiation, 273–5
Trichinella spiralis, 268–71
  irradiation of, 270–71
Trypanosoma brucei, irradiation, 271
Trypanosoma cruzi, irradiation, 271
Trypanosoma gambiense, irradiation, 271
Turkey, heat and radiation sterilized, PDR index of, 303

U

Uranium-235, 61–3, 66, 68
Uranium-238, 71
U. S. Food and Drug Administration, 300–301, 324

V

Valence, 27–29
Van de Graaff Generator, 5, 75–8
Vegetables, process for radiation sterilized, 336–7
Viruses, *D* value for, 225
  diameter-dose relation, 224
  irradiation of, 223–5
Vitamins, fat soluble, irradiation, 131–34
  retention in canning and radiation preservation of food, 137
  water soluble, irradiation, 134–37

W

Water, irradiated, 103–10
    active species in, 106–7
    cage effect in, 105
    hydrogen formation in, 105
    hydrogen peroxide yield in, 104
    linear energy transfer effects in, 107
    scavenger reactions in, 105–6
  radiation treatment of, 351
Wheat, insect deinfestation of, 275–6
  irradiation, 275–6
  protein, amino acid composition of, 286
Whole body irradiation, animal responses to, 260–67
  of man, effects of, 262–67, 375–6
    protective agents against, 265
    recovery from, 262–3
  of mice, 261–2
Worms, *see Helminths*

X

X-rays, 3, 81–2
Xanthium stems, irradiation, 239–40

Y

Yeasts, irradiation, 223
  increase of ergosterol content by irradiation, 227

Z

Zirconium-Niobium, fission products, 73
  half lives, 17
  radiation processing cost analysis, 365–72
  radiation sources, cost of, 74

CENTRAL FOOD TECHNOLOGICAL RESEARCH INSTITUTE
LIBRARY
* MYSORE. *

✓ NS. 22/c0150

F.A-0

C. F. T. R. I. LIBRARY, MYSORE.

Acc No. 5840 F8,3:6;R N60

Call No. ~~F81,3 2P,F1~~ ~~N60~~

Please return this publication on or before the last DUE DATE stamped below to avoid incurring overdue charges.

Due Date	Return Date	Due Date	Return Date
4-2-77	2/8/1		
~~Ret. 133~~			
P-230			
29/4/96	29/5/96		
2.11.04	19.10.04		
17/6/05	18/6/05		
13-11-05	11.11.05		

RIFIED
2013

CFTRI-MYSORE
5840
Radiation techno.

FAO
~~CP~~
~~693: (66)~~ ~~KO~~
OSIER(N. W)
Tech-